SEWAGE SLUDGE AS A SENSITIVE INDICATOR FOR AIRBORNE RADIONUCLIDES FROM NUCLEAR POWER PLANTS

Studies of distribution and behaviour of activation products in the terrestrial environment

Tor Ingemansson

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Abstract

Sewage sludge collected at waste water treatment plants located in the vicinity of nuclear power stations, has been shown to be a sensitive and convenient indicator for airborne locally released activation products, ^{60}Co, ^{65}Zn, ^{58}Co and ^{54}Mn. We have therefore been able to study the distribution and behaviour of these radionuclides in the terrestrial environment of three Swedish nuclear power stations. Comparative measurements on ground level air and on samples of lichen (Cladonia alpestris) and soil have also been performed. The variation by distance from the power station of ^{60}Co measured in sludge as well as on air-filters could be described by the same power function. The temporal variation of the activity concentration in sludge samples well reflects the variation of the reported release rate of airborne radionuclides from the power stations if the prevalent wind direction is taken into consideration. The relation between the activity ratio $^{60}Co/^{7}Be$ in air and in sludge was investigated and indicated that most of the detected ^{60}Co and part of ^{58}Co and ^{54}Mn activity is released from a local source and is dry deposited on the ground before it is washed off by rain.

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Sewage sludge, radioecology, airborne releases, bioindicators, atmospheric dispersion, distribution pattern, radioactive aerosols, activation products.

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These papers will be refered to in the text by their Roman numerals given above.

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1 INTRODUCTION

Radioactive material emanating from ^{238}U, ^{235}U and ^{232}Th, and their decay products, together with ^{40}K and radionuclides continuously produced through nuclear reactions in the atmosphere by cosmic radiation have always been present in our environment.

Atmospheric tests of nuclear weapons, especially the high yield tests during 1956-1958 and 1961-1962 in the northern hemisphere, have introduced a large number of different radionuclides into our environment. The main part of these radionuclides is now almost completely deposited on the ground and is now present in soil, plants, sediment, water, fish, animals and man (Persson,1970; Mattsson,1972; Carlsson, 1976; Holm, 1977). During the last two decades most atmospheric tests of nuclear weapons in the northern hemisphere have been performed in The People´s Republic of China. These tests have introduced some "fresh" radioactive debris into the biosphere.

To this globally distributed radioactive material is added radionuclides released into the biosphere from local sources. Such sources may be hospitals where radionuclides are used for diagnosis and treatment of patients. These medically used radionuclides are mostly released to the community waste water systems via patient excreta (Sodd et al.,1975; Moss,1973 ;Erlandsson and Mattsson, 1978).

Other sources of increasing significance are the nuclear

industry, reprocessing plants and nuclear power stations which release radioactive material into the cooling water as well as to the air. The releases to the marine environment are dominated by the activation products ^{60}Co, ^{65}Zn, ^{58}Co, ^{51}Cr and $^{110}Ag^{m}$ (Nilsson, 1981; Grimås,1979). The gaseous radioactive releases from nuclear power reactors are dominated by inert gases,most of them with short half lives, which are rapidly dispersed in the atmosphere after their release (Pendergast, 1979; Draxler, 1980). Inert gases with long half lives, e.g. ^{85}Kr, $^{131}Xe^{m}$ and ^{133}Xe contribute to the global atmospherical inventory of radionuclides. Besides the inert gases, fission and activation products in particulate form are also released to the air. In this thesis studies of activation products released to the air from nuclear power stations are reported.

It has been found that these locally distributed, airborne radionuclides can be studied conveniently by measurements on sewage sludge collected at treatment plants for waste water. Besides measurements on sludge, comparative measurements on samples of ground level air and on lichen (Cladonia alpestris) and soil have also been carried out.

2 PURPOSE OF THE PRESENT INVESTIGATION

This study is related to the three Swedish nuclear power stations at Simpevarp (Oskarshamn), Ringhals and Barsebäck. The airborne radioactivity released from the power stations has been the subject of theoretical studies

(Stenquist et al., 1975; Gyllander et al., 1976; Haeggblom et al., 1966). The purpose of the present investigation is to study the usefulness of sewage sludge as an indicator of airborne radionuclides both for qualitative and quantitative studies. It is hoped that these measurements will give information on the distribution and behaviour of activation products released to the terrestrial environment by nuclear power stations. The experimental results are compared with well-known transport models.

3 SOURCES, PRODUCTION AND PATHWAYS OF AIRBORNE ACTIVATION PRODUCTS

All sampling has been carried out in the surroundings of the three Swedish nuclear power stations at Simpevarp (Oskarshamn), Ringhals and Barsebäck. All three power stations are located in the southern part of Sweden. Data on the reactors at the different power stations are shown in Table 1.

The main local sources of the activation products studied are the boiling water reactors (Table 1). The contribution from the pressurized water reactors to the air of these radionuclides is comparatively small. All the studied activation products are produced by neutron-induced nuclear reactions in corrosion products deposited on the surface of the fuel elements. In Figure 1 are shown the possible pathways from the reactor to the air and the typical flow rates of gases in the different systems for a 500 MW(el) reactor. The principle design of a monitoring

TABLE 1 Some technical data for the Swedish nuclear power reactors in operation or under construction on January 1st, 1982.

Reactor	Name	Type	In operation	Thermal effect (MW)	Electric effect (MW)	Stack height (m)	Delay time of off gases
Simpevarp	01	BWR	1971	1375	440	70	30 min
	02	BWR	1974	1750	580	110	60 min
	03+	BWR		3150	1060		
Ringhals	R1	BWR	1975	2270	750	110	35 min
	R2	PWR	1975	2440	800	45	1-45 d
	R3	PWR	1980	2780	915	45	1-45 d
	R4+	PWR		2780	915		
Barsebäck	B1++	BWR	1975	1700	580	110	10 h
	B2++	BWR	1975	1700	580	110	10 h
Forsmark	F1+++	BWR	1980	2700	900	110	20 h
	F2+++	BWR	1981	2700	900	110	20 h
	F3+	BWR		300	1050		

+ Under construction.

++ Equipped with catalytic recombiner.

+++ Equipped with catalytic recombiner and inert gas separator.

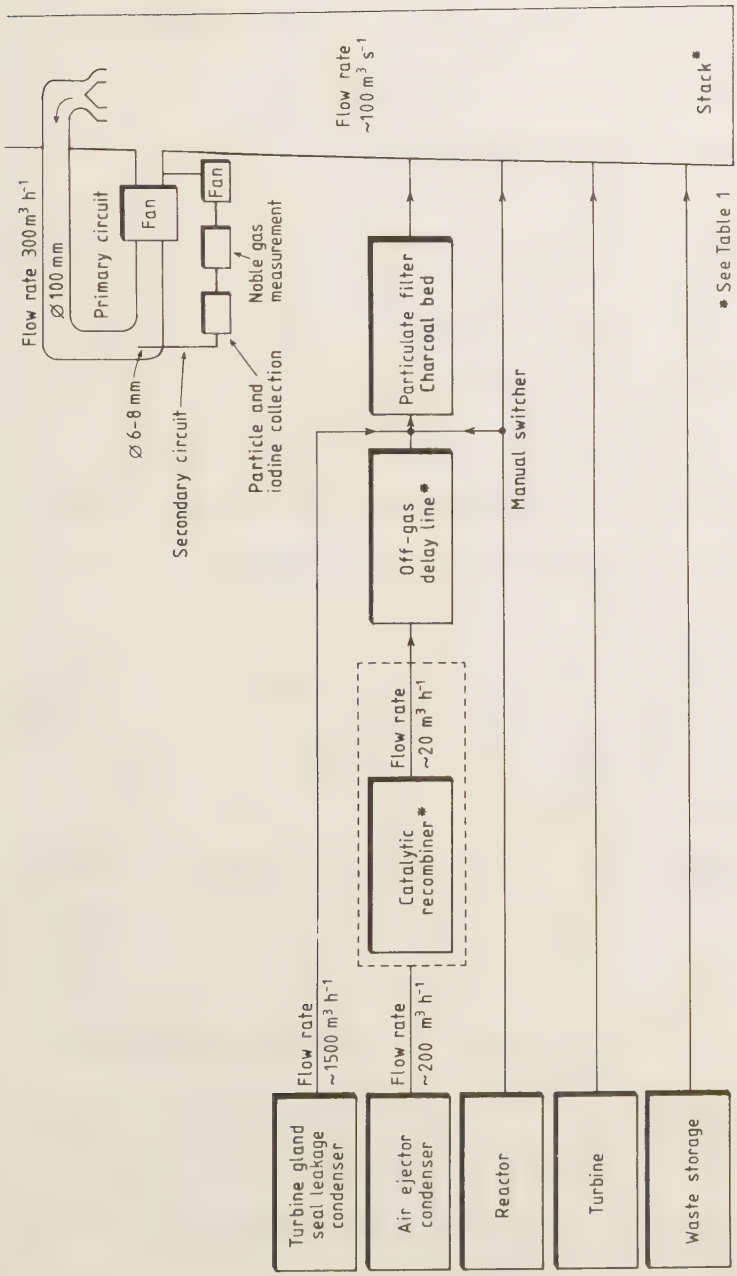

FIGURE 1. Pathways for the gaseous radioactive releases from a 500 MW(el) boiling water reactor. The principal design of a radionuclide discharge monitoring system is also shown.

system, used for measurements on released activity, is also shown.

4 INDICATORS AND MEASUREMENTS

Three different forms of samples were collected, sewage sludge from waste water treatment plants, ground level air, lichen (Cladonia alpestris) and soil. The samples were analysed with the aid of large volume and high energy resolution Ge(Li) detectors placed in 8-10cm thick lead caves. By using γ-spectrometric methods it was possible to determine the presence of several radionuclides simultaneously without destroying the samples.

Almost all sludge samples have been dried at 105°C for 24h. The detection limit (corresponding to 3 S.D. of the number of background counts) for a dried sample (0.1 kg in a 180-ml plastic tub) was about three times lower than for an untreated sample (2 kg in a 2 litre plastic bottle). The escape of radionuclides during the drying process was investigated. Two samples were collected from a well mixed amount of digested sludge. One was measured without any drying treatment, the other was dried at 105°C for 24h before it was measured. The result of the analysis showed that there was a significant loss in the dried sample of one single radionuclide, ^{140}Ba from "fresh" bomb debris. The experiment was repeated several times and no loss of any other radionuclide discussed in this work was observed.

The incoming waste water to a sewage treatment plant consists mainly of domestic sewage, industrial sewage and rain run-off from the drained areas, roofs, streets etc. served by the sewage treatment plant. The mutual relation between the different sources varies from plant to plant but on the whole the contribution from each source can be estimated to be about one third. Airborne pollution is either washed out of the atmosphere by precipitation and is then wet deposited or dry deposited on the ground. In both cases the precipitation may transport more or less of the deposited material to the sewage plant. Therefore a sample of sewage sludge reflects the deposition averaged over a large area, which is a considerable advantage relative to samples collected from small areas (0.25-5 m^2). The ratio between the estimated drained area, served by a sewage plant and the daily amount of dry sludge produced varies between 300 and 3000 $m^2.kg^{-1}$ for the different sewage plants. Therefore a sample of dry sludge (~ 0.1kg) normally reflects what is washed off from an area of 30 to 300 m^2. This may explain the extreme sensitivity of sludge as an indicator.

5 RESULTS AND DISCUSSION

From the results of this investigation it is evident that the measured ^{60}Co activity in sludge, ground level air, lichen and soil is released to the air from the nuclear power stations. It must be stressed that these releases are small and the measured ^{60}Co activity in all kinds of

samples is small relative to the measured activity of naturally occuring or man-made globally distributed radionuclides. The small releases, however, in combination with the advanced analytical techniques available are extremely useful for studies of the distribution and behaviour of radionuclides in the terrestrial environment.

Sludge was found to be the most sensitive and convenient of the indicators used. Table 2 shows the man-made and cosmogenically produced gamma-emitting radionuclides detected in sludge, air filters or in biomass samples. Those radionuclides which are present in the small reported releases to the air from the power stations, with relevant physical data are marked in the table. In this study the most extensively investigated radionuclides are the activation products ^{60}Co, ^{58}Co, ^{65}Zn and ^{54}Mn. The activation products detected in sewage sludge can be arranged in the following activity concentration sequence: $^{60}Co > ^{65}Zn > ^{58}Co \geq ^{54}Mn$ (II). During the whole sampling period, February 1978-December 1981, the activity concentration of different radionuclides in ground level air and precipitation over southern Sweden at several locations was also available (Arntsing, 1978-1981; Vintersved, 1978-1981). These measurements are carried out in a national network of sampling stations, which are all located far from the nuclear power stations (Vintersved et al., 1982). ^{65}Zn, ^{58}Co and ^{54}Mn are found in small amounts in the ground level air in connenction with atmospheric tests of nuclear weapons. There are only a few investigators who have reported ^{60}Co activity in the

TABLE 2 Man-made and naturally occuring gamma-emitting radionuclides detected in samples of sludge, air filters, lichen and soil with relevant physical data. The radionuclides present in the reported releases (r.r.) to air from the nuclear power stations are marked.

Nuclide	Main production mechanism f=fissionprod.	Half life	Dominating gamma-ray energy (MeV)	intensity per 100 decays	Present in r.r.to air	Detected in sludge	air filters	lichen & soil
^{7}Be	spallation	53.3d	0.478	10.4		x	x	x
^{22}Na	spallation	2.6y	1.275	99.9			x	
^{51}Cr	$^{50}Cr(n,\gamma)$	27.7d	0.320	10.2	x	x		
^{54}Mn	$^{54}Fe(n,p)$	312d	0.835	100	x	x	x	x
^{57}Co	$^{58}Ni(n,2n)^{57}Ni(\beta^{+})$	272d	0.122	85.6		x		
^{58}Co	$^{58}Ni(n,p)$	70.8d	0.811	99.4	x	x	x	
	$^{59}Co(n,2n)$							
^{60}Co	$^{59}Co(n,\gamma)$	5.27y	1.173	100	x	x	x	x
			1.332	100				
^{65}Zn	$^{64}Zn(n,\gamma)$	244d	1.116	50.8	x	x	x	x
^{75}Se	$^{74}Se(n,\gamma)$	119d	0.136	54		x		
			0. 65	58				

TABLE 2 Cont.

Nuclide	Main production mechanism	Half life	Dominating gamma-ray energy (MeV)	Dominating gamma-ray intensity per 100 decays	Present in r.r.to air	Detected in sludge	Detected in air filters	Detected in lichen & soil
^{88}Y	f	107d	1.836	99.3			x	
^{95}Zr	$^{94}Zr(n,\gamma)$, f	64.0d	0.724	44.3	x	x	x	
			0.757	54.6				
^{95}Nb	$^{95}Zr(\beta^-)$, f	35.0d	0.766	99.8	x	x	x	
^{99}Mo	$^{98}Mo(n,\gamma)$, f	66.0h	0.739	12.6				
$^{99}Tc^m$	$^{99}Mo(\beta^-)$ (87%)	6.0h	0.141	89.0	x			
^{99}Tc ↓	$^{99}Mo(\beta^-)$ (13%)	2.1×10^5y						
^{103}Ru	f	39.4d	0.497	86.4		x	x	
^{106}Ru	f	367d						
^{106}Rh	$^{106}Ru(\beta^-)$	29.8s	0.622	9.8		x	x	x
$^{110}Ag^m$	$^{109}Ag(n,\gamma)$	252d	0.658	94.4		x		
^{125}Sb	$^{125}Sn(\beta^-)$, f	2.7y	0.428	30		x	x	x
^{131}I	f	8.05d	0.364	81	x	x	x	
^{134}Cs	$^{133}Cs(n,\gamma)$, f	2.06y	0.605	97.6		x	x	
			0.796	85.4				

TABLE 2 Cont.

Nuclide	Main production mechanism	Half life	Dominating gamma-ray energy (MeV)	intensity per 100 decays	Present in r.r.to air	Detected in sludge	air filters	lichen & soil
^{137}Cs	f	30.2y	0.662	85	x	x	x	x
^{140}Ba	f	12.8d	0.537	23.6	x	x	x	
^{140}La	$^{140}Ba(\beta^-)$, f	40.3h	0.487	43	x	x	x	
			1.596	96				
^{141}Ce	f	32.6d	0.145	48.0		x	x	
^{144}Ce	f	285d	0.134	11.1		x	x	x
^{144}Pr	$^{144}Ce(\beta^-)$, f	17.3min	0.696	1.3				
^{155}Eu	$^{154}Sm(n,\gamma)$ $^{155}Sm(\beta^-)$	4.96y	0.105	23			x	
^{241}Am	$^{241}Pu(\beta^-)$	432y	0.060	35.7				x

terrestrial environment or in ground level air, which owes its origin to global fallout (Roy et al., 1981a, 1981b; Arntsing, 1978-1981) and then only in very small amounts and in single samples. The contribution from global fallout of ^{60}Co activity to the sludge, to the ground level air or to the lichen and soil samples seems therefore to be of no significance for the results of this work. Because of this and the fact that the ^{60}Co concentration was always much greater than that of the other activation products in all kinds of samples, this radionuclide has been used both for qualitative and quantitative studies.

From measurements on sludge (I and IV) and on air filters (III) it can be deduced that the distribution, D(x), by distance, x, from the power stations can be described by the power function

$$D(x) = \begin{cases} \frac{c}{x\,\sigma_z} \exp\{-h^2/2\sigma_z^2\} & x \leq 8 \text{ km} \quad (1) \\ \alpha x^{-2.5\pm0.5} & 8 \text{ km} < x < 52 \text{ km} \quad (2) \end{cases}$$

where c and α are constants, h(m) is the effective chimney height and σ_z (m) the vertical dispersion coefficient. σ_z is dependent on the prevalent weather condition (Pasquill's A-F categories, A is the most turbulent and F the most stable weather condition), and the distance from the release point. Values of σ_z can be obtained from a graphical representation (Slade, 1968) or can be easily evaluated for any distance or weather condition using the expression (Wash 1400, 1975)

$$\ln\sigma_z = \ln c_1 + c_2 \ln x + c_3 (\ln x)^2 \qquad (3)$$

c_1, c_2 and c_3 are parameters associated with each weather type and are obtained by curve fitting under representative dispersion conditions. The neutral weather type, Pasquill´s D-category is prevalent for about 60% of the time over southern Sweden (Finck et al., 1979). As the measured activities in the air filters are mean values over long time periods (6-7 days), this weather type has been assumed in all calculations. For Pascuill´s D-category c_1=0.036, c_2=1.24 and c_3=-0.0248 (Wash 1400, 1975).

The activation products are produced by neutron capture (Table 2) outside the fuel elements. Because of the long physical half life of ^{60}Co (5.3y) it seems most likely that the release of this radionuclide will increase during the first ten years of operation of a reactor because of the build up of activity. After a time the build up and decay of the ^{60}Co activity will reach equilibrium.

The fission products are produced inside the fuel elements. Because of small faults in the cladding material some activity can leak into the primary coolant water. Small amounts of long-lived particulate fission products are also present in the airborne releases from the power stations. It cannot be excluded that these radionuclides may be deposited in the vicinity of the power stations. Because of the large amount of ^{137}Cs activity from global fallout, already present in the air as well as on the

ground, it has not been possible to determine from the indicators used if small local releases make any significant contribution to the terrestrial environment.

6 CONCLUSION

It has been shown that sewage sludge collected at treatment plants for waste water is an extremely sensitive and convenient indicator for locally released airborne activation products. The sludge reflects very well the variations of the reported release rate to the air from the power stations. Through measurements on sludge it is also possible to study the distribution and behaviour in the terrestrial environment of radionuclides from local sources or global fallout.

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SEWAGE SLUDGE—A POSSIBLE INDICATOR FOR RADIONUCLIDES RELEASED TO THE ATMOSPHERE FROM NUCLEAR POWER PLANTS

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Abstract—Activation products typical of the release from nuclear power stations have been detected in sludge collected in treatment plants for community waste water located between 2 and 52 km from the Swedish nuclear power stations at Ringhals and at Simpevarp near Oskarshamn. The time variations of the ^{60}Co activity concentration at two sampling sites located in the same direction from the Oskarshamn nuclear power station have been followed during 5 months and found to be almost identical. For distances, z, between 8 and 52 km from the nuclear power stations, the ^{60}Co/^{7}Be activity ratio, $R(z)$, decreases rapidly and the variation with distance follows the power function $R(z) = \alpha \cdot z^{-\beta}$, where α and β are constants. The β-values for the Ringhals and Oskarshamn areas were not significantly different and their mean value was 2.5 ± 0.5. Possible sources of the activation products in sewage sludge and the transport mechanism to the community treatment plants are discussed.

1. INTRODUCTION

We have earlier shown sewage sludge to be a very sensitive indicator for radioactive materials released from hospitals (Er78) or deposited on the ground after tests of nuclear weapons in the atmosphere (Ma79). In these studies, samples of sewage sludge were also collected at community waste-water treatment plants in the surroundings of the Swedish nuclear-power stations at Ringhals and at Simpevarp near Oskarshamn. A number of activation products were detected in these samples. The purpose of this study was to try to map the distribution of ^{60}Co around the power plants and to obtain information on the transport of the radionuclides from the power stations to the sewage treatment plants.

2. MATERIAL AND METHODS

The Oskarshamn nuclear-power station at Simpevarp is situated on the east coast of Sweden (57.4°N, 16.7°E) and has two boiling-water reactors with 1375 and 1750 MW thermal effect. They were placed in operation in 1971 and 1974 respectively. The locations and the types of the six sewage treatment plants around the Oskarshamn nuclear power station are shown in Fig. 1 and Table 1. Sampling was performed during the periods February–May 1978 and September 1978–January 1979. At three of the plants, Figeholm, Oskarshamn and Hultsfred, the sludge was sampled at approx. 1-week intervals during a 5-month period.

The Ringhals nuclear power station on the west coast of Sweden (57.3°N, 12.1°E) has one boiling-water and one pressurized water reactor with thermal effects of 2270 and 2440 MW respectively. Both reactors became operational in 1975. The locations and types of nine sewage treatment plants used in studying the distribution of activation products

FIG. 1. Map showing locations of the nuclear stations at Ringhals and Simpevarp (Oskarshamn) (■) in Sweden and the community waste water treatment plants used for collection of sewage sludge (●). Wind-roses at a height of 100 m for the power stations are also presented.

around this nuclear power station in the period June–September 1979 are also given in Table 1 and Fig. 1. In this table information is also given about the treatment processes of the different plants. Most of them have both mechanical, biological and chemical steps. The final treatment of sludge is done by dehydration in a centrifuge or a press. For some of the plants this step is omitted. The fraction of the total amount of incoming waste water which is run-off rainwater varies considerably from the plant to plant but as a mean over the year the rainwater constitutes about half of the waste water. All samples of sewage sludge were collected at the final output of the treatment plant.

All the samples of sewage sludge collected during February–May 1978 were packed into 2000 ml plastic containers without any treatment . All other samples collected at different sewage treatment plants were dried at 105°C for 24 hr. The dried samples (≈ 100 g) were homogenized and packed in 180-ml plastic containers. The measurements were carried out using Ge(Li)-detectors of 46–100 cm^3 volume placed in 8-cm-thick lead caves in a low-background laboratory. The counting times varied between 15 and 72 hr per sample. The efficiencies of the detectors were carefully determined using calibration samples of ^{152}Eu, ^{57}Co and ^{22}Na with known activities. All the activity concentrations given in this paper have been corrected for decay to the time of collection.

3. RESULTS AND DISCUSSION

The activation products detected in sewage sludge are $^{60}Co(T_{1/2} = 5.27a)$, ^{65}Zn $(244d)$, $^{58}Co(70.8d)$, $^{54}Mn(312d)$, $^{57}Co(272d)$, $^{110}Ag^m(252d)$ and $^{51}Cr(27.7d)$. All these radionuclides occur frequently in the reactor water and liquid releases from the nuclear power stations (Ma80, Ni80). In Fig. 2 the time variation of the ^{60}Co activity concentrations in sewage sludge (dry weight) from Oskarshamn and Hultsfred during February–May 1978 and from Figeholm, Oskarshamn and Hultsfred during September 1978–January 1979 are shown. Comparison of the activity concentrations of ^{60}Co at Figeholm

Table 1. Collecting sites for sludge in the surroundings of two Swedish nuclear power stations

Site number	Name	Type[+]	Distance from the power plant (km)	Incoming water (m^3/d)	Sludge output dry weight (kg/d)	Mean residence time for sludge (d)
Simpevarp area:						
1	Figeholm	m,b,c,t	8.2	1 000	100	3 - 5
2	Oskarshamn	m,b,t	21	11 000	1 900	3 - 4
3	Kristdala	m,b,c,t	27	1 000	100	3 - 5
4	Ankarsrum	m,b,c,t	38	1 000	100	3 - 5
5	Västervik	m,b,c,t,d	39	13 000	2 300	32 - 36
6	Hultsfred	m,b,c,t	52	4 000	750	3 - 5
Ringhals area:						
11	Bua	m,b,c,t	2.4	1 500	300	3 - 5
12	Askloster	m,b	9.0	550	60	6 - 7
13	Veddige	m,b	13	1 500	170	6 - 7
14	Varberg	m,b,c,t,d	19	11 000	2 300	40 - 45
15	Horred	m,b	25	440	150	3 - 5
16	Björketorp	m,b	31	280	70	4 - 5
17	Skene	m,b,c,t	44	7 000	1 500	4 - 5
18	Falkenberg	m,b,c,t	46	10 000	4 000	3 - 6
19	Borås	m,b,c,t,d	75	50 000	8 600	24 - 27

[+] m = mechanical step,
b = biological step,
c = chemical step,
t = sludge dehydrated in press or centrifuge,
d = sludge digester

and Oskarshamn shows an almost identical time-variation pattern. This indicates that the source of the measured ^{60}Co-activity is the same and that it may be spread via the air. The activity concentrations of various radionuclides in sludge collected at different sites vary strongly. These differences may depend on the various distances from the source but also on the different proportions of rain water in the waste water reaching the different sewage treatment plants. In previous work (Ma79), it was shown that for radionuclides recently deposited on the ground, these differences can be eliminated by dividing the activity concentration of the radionuclide of interest by the activity-concentration of the cosmic-ray-produced radionuclide ^{7}Be($T_{1/2} = 53.3d$).

Figure 3 presents the ^{60}Co/^{7}Be activity ratio in sewage sludge from waste-water plants at various distances from the Oskarshamn nuclear power station during March–December 1978. The various sampling sites are indicated at the bottom of the diagram by site numbers given in Fig. 1 and Table 1. The figure shows that the decrease in the activity ratio, $R(z)$, with distance, z, from the nuclear power station may to a first approximation be described by the power function

$$R(z) = \alpha \cdot z^{-\beta} \quad (\text{for } 8\text{ km} < z < 52\text{ km}) \tag{1}$$

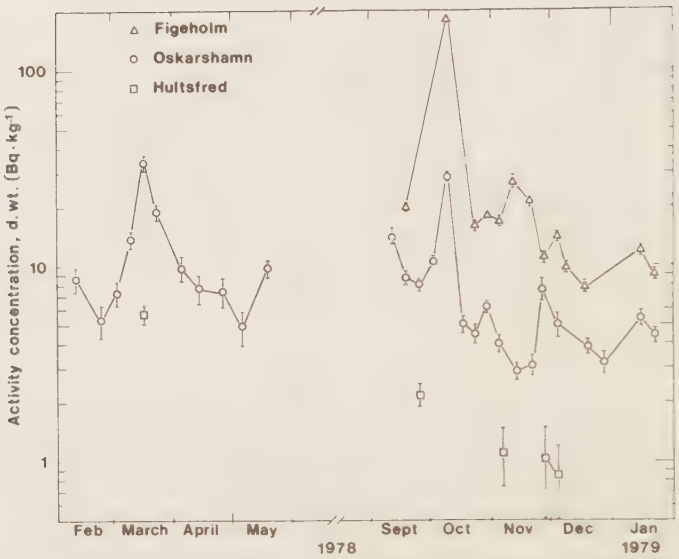

FIG. 2. Time variation of the ^{60}Co activity concentration (dry weight) in sewage sludge collected in Oskarshamn (○), Figeholm (△) and Hultsfred(□).

where α and β are constants. The mean of the β-values at various times calculated for the Oskarshamn area is 2.3 ± 0.5 (S.D.). The same types of activation products as were found around the Oskarshamn nuclear power station were also found in the surroundings of Ringhals.

Figure 4 shows that the variation with distance from Ringhals of the ^{60}Co/^{7}Be activity ratio in sewage sludge from waste water treatment plants can also be described by equation (1). The collecting sites for sewage sludge around the Ringhals nuclear power station are located in two main directions, NE and SE, relative to the power station. When calculating the β-values in the Ringhals area, these two directions were treated separately. No significant difference in the β-values was found, however. The β-value for the Ringhals area in July–September 1979 was found to be 2.6 ± 0.7 (S.D). The differences in the β-values for the Oskarshamn and Ringhals environments are not significant and both are consistent with an inverse-square behaviour which would be expected if the released activity is distributed isotropically.

The origin of the activation products found in sewage sludge from the areas around the nuclear power stations is not definitely established but the results of this work strongly suggest that these power stations can be the sources. Furthermore, the similar time variation in Figeholm and Oskarshamn for the activity concentration of ^{60}Co and the similar profiles for the activity ratio ^{60}Co/^{7}Be around the two nuclear power stations indicate that a probable way for the transport of the activation products is via the air. Indeed this is the most likely transport

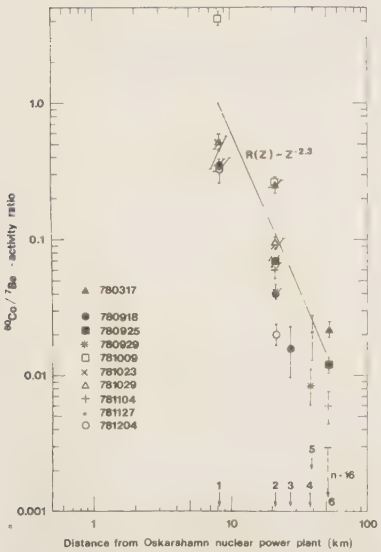

FIG. 3. ^{60}Co/^{7}Be-activity ratio in sewage sludge from waste water treatment plants located at different distances from the nuclear power station at Simpevarp (Oskarshamn). The figures at the bottom of the diagram are the site numbers of Table 1 and Fig. 1. Dates of measurement are given at the left.

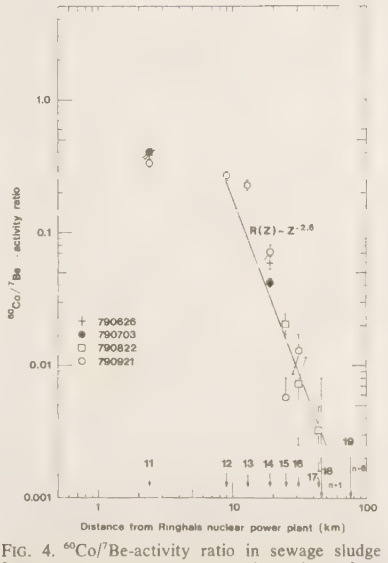

FIG. 4. ^{60}Co/^{7}Be-activity ratio in sewage sludge from waste water treatment plants located at different distances from the nuclear power station at Ringhals. The figures at the bottom of the diagram are the site numbers of Table 1 and Fig. 1.

mechanism, as the general direction of water flow in both areas would not permit transport from the presumed sources to the detection sites (Fig. 1).

In order to determine whether traces of deposited ^{60}Co could be found also on the ground, samples of lichen (*Cladonia alpestris*) were collected at different distances from the nuclear power station at Simpevarp in September and November, 1979. The locations were chosen along a radius in a direction 225° from the plant. The September samples were collected from 0.25 m^2 down to a depth of 2–3 cm, dried at 105°C for 24 hr and then measured in 180 ml plastic containers in the same manner as the sewage-sludge samples. Significant amounts of ^{60}Co were found in some of the lichen samples and showed decreasing concentration with distance from the plant (Table 2).

In November, one additional 5-m^2 lichen sample was taken at a distance 6.2 km from the Simpevarp plant. The sample was dried as above and then ashed at 600°C to increase the sensitivity of the measurements. The ^{60}Co area content in the lichen carpet was (0.16 ± 0.01) Bq · m^{-2}.

The lichen measurements cannot be used for quantitative deposition estimates because they are too few and the uptake and retention patterns of the deposited ^{60}Co activity in the relatively thin lichen carpets are not known. The findings however strongly indicate that the ^{60}Co and other activation products detected in the sewage sludge are released to the

Table 2. Content of ^{60}Co in carpets of lichen (Cladonia alpestris) *from the surroundings of the nuclear power station at Simpevarp near Oskarshamn. The thickness of the lichen carpet was* (1.60 ± 0.18) kg d.wt per m^2

Date	Distance from power plant km	^{60}Co area content Bq · m^{-2} (± S.D)
1 Sept. 1979	0.9	5.8 ± 1.2
"	2.1	0.79 ± 0.32
"	4.1	1.2 ± 0.6
"	5.2	0.58 ± 0.30
"	6.2	0.44 ± 0.20
"	7.0	< 0.2
"	9.0	< 0.2
"	10	0.37 ± 0.21
11 Nov. 1979	6.2	0.16 ± 0.01*

* ashed sample 5 m^2

atmosphere from the power plants and then deposited on the ground. Another explanation could be fallout from tests of nuclear weapons or releases from nuclear power plants outside Sweden, but the activity ratio $^{60}Co/^{7}Be$ in precipitation and ground-level air has not exceeded 2.5×10^{-4} and 1.2×10^{-4}, respectively, during recent years. These measurements have been performed by the National Institute of Radiation Protection (Vi80) at various other locations in Sweden.

In Table 3 the total monthly "outputs" of ^{7}Be and ^{60}Co via the sludge of four wastewater treatment plants are presented. The total area of the town of Oskarshamn is 30 km^2 but the estimated drained area is only 2.8 km^2. Using equation (1) for distances between 8 and 50 km and a constant deposition up to 8 km it is possible to estimate a minimum release of ^{60}Co from the nuclear power plant. If all the ^{60}Co activity detected is airborne and has an isotropic distribution, a uniform deposition velocity over all types of terrain, and if all the deposited ^{60}Co activity is transferred to the sludge the atmospheric release of ^{60}Co during 1978 must have been around 15 GBq (0.4 Ci) for the Oskarshamn nuclear power station.† The release rate of ^{60}Co to the atmosphere reported was however only 0.06 GBq per year (Gr80). This means that our estimated release is 250 times higher than reported.

The investigation in the Ringhals area has been too short to enable the yearly release to be calculated, but the lower $^{60}Co/^{7}Be$ activity ratio in the Ringhals area relative to that in the Oskarshamn area at the same distance indicates that the release of ^{60}Co from Ringhals is 30–60% lower than that from Oskarshamn.

Our calculation of the release of ^{60}Co can be affected by the following factors:

(1) The transport of ^{60}Co from the air to the sludge can be divided into two com-

†The annual ^{60}Co deposition for Oskarshamn was determined by using the average monthly activity release found in the sludge (0.56 MBq/month from Table 3), multiplying by 12 for the entire year and dividing by the relevant deposition area (2.8 km^2). This gives a deposition rate of about 2.4 MBq km^2 yr^{-1} for the vicinity of Oskarshamn. When the value $\beta = 2.3$ is used in equation (1), α is determined to be $\alpha = 2.6 \times 10^3$. Using these parameters in equation (1) determines the deposition rate at an 8-km radius from the power plant to be 22 MBq km^{-2} yr^{-1}. Simple integration of the deposition rate over the two areas gives the result 15 GBq/yr.

Table 3. Estimated total activity of ^{7}Be and ^{60}Co transported via the sludge, per month

Treatment plant for waste water	Activity release rate (MBq · month^{-1}) Feb.-May 1978 ^{60}Co	^{7}Be	Sept.78-Jan.79 ^{60}Co	^{7}Be	July 79 ^{60}Co	^{7}Be
Figeholm	+	+	0.079	0.11	+	+
Oskarshamn	0.67	6.3	0.44	6.5	+	+
Varberg	+	+	+	+	1.1^{++}	21^{++}
Bua	+	+	+	+	0.19	0.44

$^{+}$ Sampling not performed.

$^{++}$ Corrected for physical decay during 40 days.

ponents. The first is the ^{60}Co washed out of the air by rain and then transported to the sewage plant. The second is a slower process where the ^{60}Co is first deposited on the ground, houses or other obstacles and then washed off by rain-water and transported to the plant. It is most likely that both these processes can be correlated to the ^{7}Be activity.

(2) An investigation of the use of radioactive material of the industrial establishments and of the hospital in the town of Oskarshamn has shown that none of these facilities have used or handled ^{60}Co.

(3) The effective area washed by rain is difficult to estimate and may differ from the 2.8 km^2 which was given by the authorities of Oskarshamn and which was used in our calculations. The total amount of incoming water to the sewage treatment plant is 4.0×10^6 m^3 year^{-1} (Table 1). According to the staff at the treatment plant about one third of the incoming water is rain runoff, one third domestic sewage and one third industrial sewage or 1.3×10^6 m^3 yr^{-1} each. The annual precipitation over Oskarshamn is 600 mm. To get 1.3×10^6 m^3 of rainwater per year the precipitation from 2.2 km^2 has to be collected. This area is in good agreement with the estimated 2.8 km^2.

(4) It is also possible that some of the ^{60}Co activity reaches the sewage system from food and/or domestic water supply. However, even if all domestic water consists of pure rain water this should give a total effective drained area of 6.6 km^2, which could reduce our factor of 250 between estimated and reported releases to a factor of about 80. It is, however, most unlikely that the domestic water supply consists of pure unfiltered rainwater and other investigations show that the ^{60}Co activity in food products is below the detection limit. We therefore believe that this gives an insignificant contribution to the ^{60}Co input to the waste water treatment plant.

(5) The wind directions in the Oskarshamn area are rather uniform for the year as the wind-rose indicate (Fig. 1); winds from the sea are not as frequent as one would expect from the location of the power plant and we see no topographic or meteorological reasons which can account for a higher deposition over this specific area. If the efficiency of washout and washoff by rain is less than 100%, our figure would underestimate the deposited ^{60}Co activity.

The large difference between the reported and calculated releases of ^{60}Co is remarkable and it is obviously of considerable interest to study the possibility of other transport mechanisms which can account for the presence of activation products in the sewage sludge collected in the surroundings of the nuclear power stations, or to determine if

there exists a hitherto unsuspected diffuse leakage of radionuclides from these power stations.

4. SUMMARY AND CONCLUSION

Activation products have been detected in samples of sewage sludge collected in the surroundings of the two Swedish nuclear power plants at Simpevarp near Oskarshamn and at Ringhals. A 5-month comparison of the ^{60}Co activity concentrations from two collecting sites in the same direction from the Oskarshamn power plant shows an almost identical time variation pattern. This indicates that the activation products have a common source. The decrease in the ^{60}Co/^{7}Be activity ratio, R, with distance, z, from the nuclear power plant follows the function $R(z) = \alpha \cdot z^{-\beta}$ $(8 < z < 52\,\text{km})$ with $\beta = 2.3 \pm 0.5$ (S.D) for the Oskarshamn area and $\beta = 2.6 \pm 0.7$ (S.D) for the Ringhals area. These findings strongly suggest that the sources of the activation products are the two nuclear power stations and that the activation products are spread via the air. The presence of ^{60}Co deposited on the ground has also been confirmed by measurements on carpets of lichen up to 6.2 km SW of the Oskarshamn nuclear power station.

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STUDIES OF ACTIVATION PRODUCTS IN THE TERRESTRIAL ENVIRONMENTS OF THREE SWEDISH NUCLEAR POWER STATIONS

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ABSTRACT

Samples of sewage sludge , lichen (Cladonia alpestris), soil and ground level air have been analysed for activation products released to the atmosphere from the three Swedish nuclear power stations at Simpevarp near Oskarshamn, Ringhals and Barsebäck. The activity concentration of the activation products in the sludge can be arranged in the following sequence: $^{60}Co > {}^{65}Zn > {}^{58}Co \approx {}^{54}Mn$. There is agreement between the time variation of the activity concentration in the sludge and the reported releases to the air from the power stations. The measured activity ratio $^{58}Co/^{60}Co$ in the sludge does not significantly differ from that reported in the releases to

the air.

The activity concentration in sludge sedimented from incoming waste water has been used to obtain better time resolution than that obtained using only digested sludge from the final step of the plant. These studies have shown that the activity concentration of ^{60}Co increases substantially with the first rain run-off that reaches the sewage plant and then falls off rapidly.

Measurements on samples of lichen and underlying soil show that the radioactive cobalt isotopes (^{58}Co and ^{60}Co) have a short mean residence time in the lichen carpet compared with most fission products present in global fall-out.

INTRODUCTION

Small amounts of radionuclides are continuously released through tall chimneys to the air from nuclear power stations during periods of normal power production. These airborne radionuclides may be deposited in the environment. Because of their very small activity, they are difficult to detect and quantify. Sewage sludge has, however, been shown to be a very sensitive indicator for radioactive material released from different sources (Mattsson et al.,1977; Erlandsson & Mattsson, 1978; Mattsson et al., 1979; Prichard et al., 1981) and strong

indications have also been found that ^{60}Co detected in sludge samples collected at waste water treatment plants in the surroundings of the two Swedish nuclear power stations at Ringhals and Simpevarp may emanate from the power stations (Ingemansson et al., 1981).

The purpose of this investigation is to extend our previous measurements to Barsebäck nuclear power station, to study other activation products than ^{60}Co also found in the sludge samples, and to compare the reported releases of the activation products to the air from the power stations with the activity concentration in the sludge samples, collected weekly in the surroundings of each power station. Comparative measurements on radioactive nuclides in samples of lichen , soil and ground level air have also been performed.

MATERIAL AND METHODS

Three different forms of samples were collected; a) sewage from waste water treatment plants b) dust on air filter papers and c) samples of lichen (*Cladonia alpestris*) and underlying soil.

All the waste water treatment plants had mechanical, biological and chemical degradation steps. At the entrance to the plant the incoming water rapidly passes first the mechanical step and then the biological step. After these steps some of the sludge was separated from the waste

water by sedimentation. The mean residence time within the plant for this material was about 1 hour. About 200 litres of the sediment was pumped into a 250 litre plastic tank during a five minute period. After 1 hour´s further sedimentation 2 litres of the sludge, with a dry substance content of about 3%, was tapped from the bottom of the plastic tank. Samples collected at this point are hereafter called "entrance sludge". After a mean residence time of three to five days the main part of the sludge reaches the dehydrator and the dry substance content is raised to 12-15%. Sludge samples were also collected after this step and are called "final sludge". The sewage treatment plants at Bua, Oskarshamn and Varberg have been described earlier (Ingemansson et al., 1981). Borgeby sewage treatment plant serves 14,000 inhabitants in the communities of Löddeköpinge and Bjärred, located 6 km east respective 6 km south-east of the power station, and some smaller villages, which lie in an area south-east to north-east, 3-10 km, of Barsebäck nuclear power station and has a daily incoming water volume of about 4200 m^3 and a daily sludge production of 1100 kg (dry weight). Entrance sludge was only collected at Borgeby. The final sludge was sampled weekly at the sewage treatment plants at Oskarshamn, 21 km south-west of Simpevarp nuclear power station during February-May 1978, September 1978-August 1979 and May 1980-January 1981, at Bua, 2.4 km south of Ringhals nuclear power station and at Borgeby, 5.8 km east of Barsebäck nuclear power station during May 1980-January 1981. All sludge samples were dried at 105°C for 24 hours, homogenized and then packed in 180 ml plastic tubs of 80

mm diameter and 35 mm height.

Ground level air was sucked through a 0.57m x 0.57m Microsorban polystyrene absolute filter placed in front of a centrifugal fan with a pumping capacity of 800 m^3h^{-1}. The filter media was 1.55 mm thick (21.7 $mgcm^{-2}$) (Suschny, 1968). The air speed through the filter was 0.7 ms^{-1}, the pressure fall over the filter was 6000 Pa and the sampling time 6-7 days. The fan was placed at Borgeby sewage treatment plant. The wind direction at the same place was carefully registered by a wind vane. The filters (about 70g) were compressed and packed in 180 ml plastic tubs.

The lichen (Cladonia alpestris) and soil, down to a depth of 5 cm, were collected from areas of 5 m^2 and 0.5 m^2 respectively at different distances and in different directions from Simpevarp in 1979 and 1980. These samples were dried at 105°C for 24 hours, ashed at 600°C and then packed in 180 ml plastic tubs.

The gamma-ray measurements were carried out using Ge(Li)-detectors with full absorption efficiencies of 10-20% (for ^{60}Co, 1.33Mev, compared with a 7.6 cm (diam.)X7.6 cm cylindrical NaI(Tl) crystal, 25 cm between the source and the detector), placed in lead caves with 10 cm thick walls. The plastic tubs with the samples were placed immediately in front of the detectors, as were the calibration samples. The efficiencies of the detectors for various photon energies were determined using samples of ^{152}Eu with known activity. All the activity concentrations

given in this paper have been corrected for physical decay back to the time of collection.

NOTE

The power station at Simpevarp, 21 km north of Oskarshamn on the Swedish east-coast has two boiling-water reactors with 1375 (O1) and 1750 (O2) MW thermal effect. They were placed in operation in 1971 and 1974 respectively. The Ringhals nuclear power station, 19 km north of Varberg on the Swedish west-coast has one boiling-water reactor and two pressurized water reactors with thermal effects of 2270 (R1), 2440 (R2) and 2780 (R3) MW respectively. R1 and R2 became operational in 1975 and R3 in 1980. Barsebäck nuclear power station is situated 18 km north of Malmö on the Swedish south-west coast and has two boiling-water reactors with 1700 MW thermal effect each. They were placed in operation in 1975 (B1) and 1977 (B2) respectively.

RESULTS AND DISCUSSION

General features of "gamma-ray spectra".

All the recorded "gamma-ray spectra" have peaks emanating from decay products of ^{238}U (^{226}Ra) and ^{232}Th and from ^{40}K and ^{7}Be, present in the samples and/or the surroundings of the detector. About 14 days after an atmospheric test of a nuclear weapon in the northern hemisphere, radionuclides

with short physical half-lives e.g. ^{140}Ba- ^{140}La, ^{131}I, ^{103}Ru and ^{141}Ce dominate the "spectra". After some time the more long lived products e.g. ^{106}Ru, ^{144}Ce, ^{137}Cs and ^{54}Mn dominate the spectra. The man-made radionuclides normally found in the ground level air are as a rule also detected in the sludge. Besides these radionuclides the sludge can contain substantial activity of ^{131}I (up to 5 $kBqkg^{-1}$, dry mass). This radionuclide is used for medical investigations and treatment of patients (Erlandsson & Mattsson, 1978; Prichard et al., 1981). The most prominent gamma-emitter in lichen is ^{137}Cs emanating from atmospheric tests of nuclear weapons.

Activation products in sewage sludge.

The activity concentration of ^{60}Co, ^{58}Co, ^{65}Zn and ^{54}Mn in weekly samples of final sludge collected at Oskarshamn, Bua and Borgeby between May 1980 and January 1981 are shown in Figures 1, 2 and 3 respectively. In the figures are given the reported weekly releases to the air from the power stations at Simpevarp, Ringhals and Barsebäck. The periods of test, production and revision for the different reactors are also presented. When the activity concentration in the sludge is below the detection limit (corresponding to less than 3 S.D. of the number of background counts), this is indicated by an arrow in the figures.

The activity concentration found in the final (as well as in the entrance sludge) can be arranged in the following

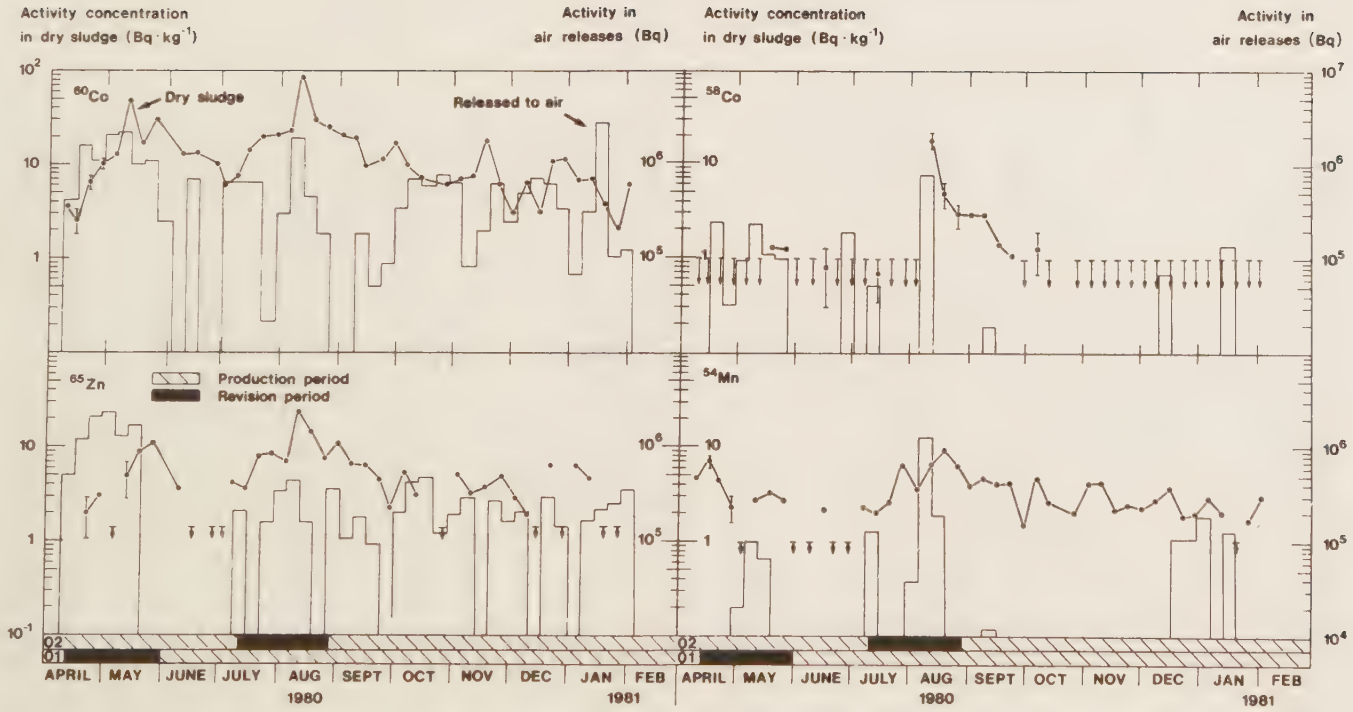

FIGURE 1 The activity concentration of ^{60}Co, ^{58}Co, ^{65}Zn and ^{54}Mn in dried samples of final sludge collected at Oskarshamn between April 1980 and February 1981. The reported weekly releases to the air from Simpevarp power station (21 km north of Oskarshamn) are also given. The production, and revision periods for the two reactors O1 and O2 are shown in the bottom of the figure.

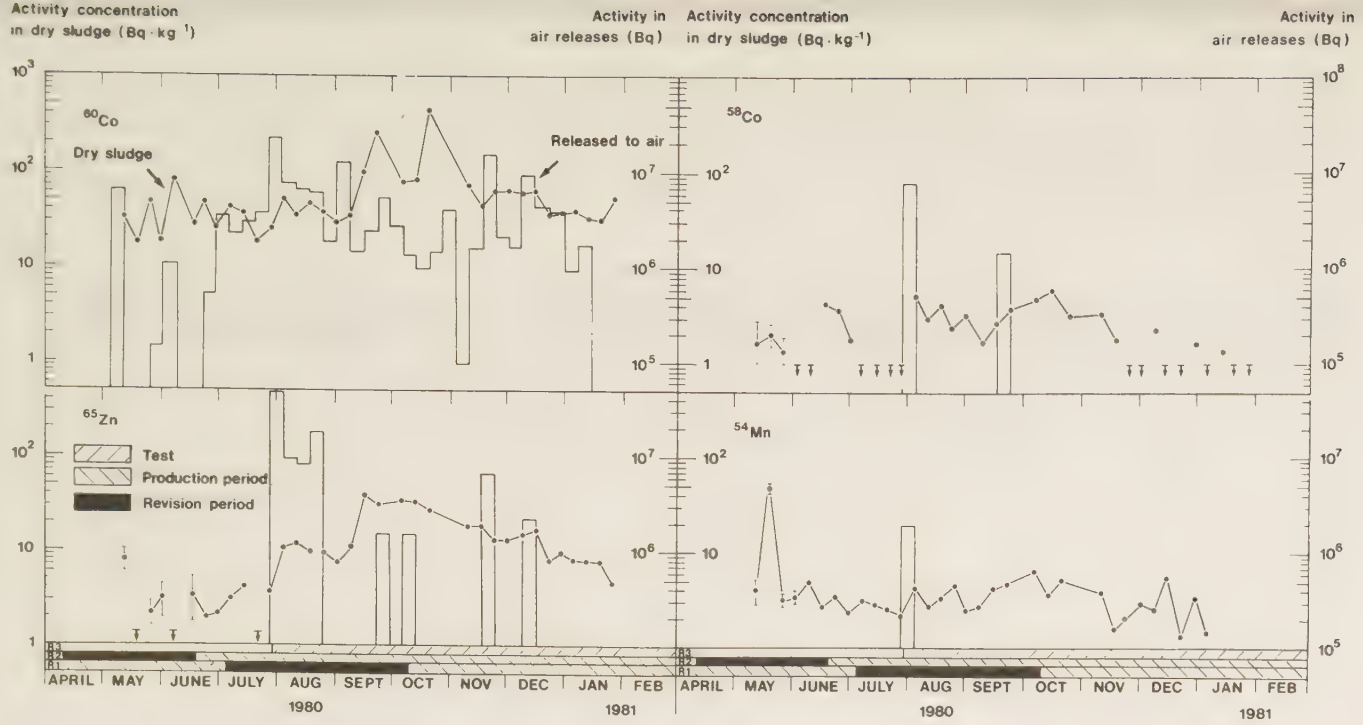

FIGURE 2 The activity concentration of ^{60}Co, ^{58}Co, ^{65}Zn and ^{54}Mn in dried samples of final sludge collected at Bua between April 1980 and February 1981. The reported weekly releases to the air from Ringhals nuclear power station (2.4 km north of Bua) are also given. The test, production, and revision periods for the three reactors (R1, R2 and R3) are shown in the bottom of the figure.

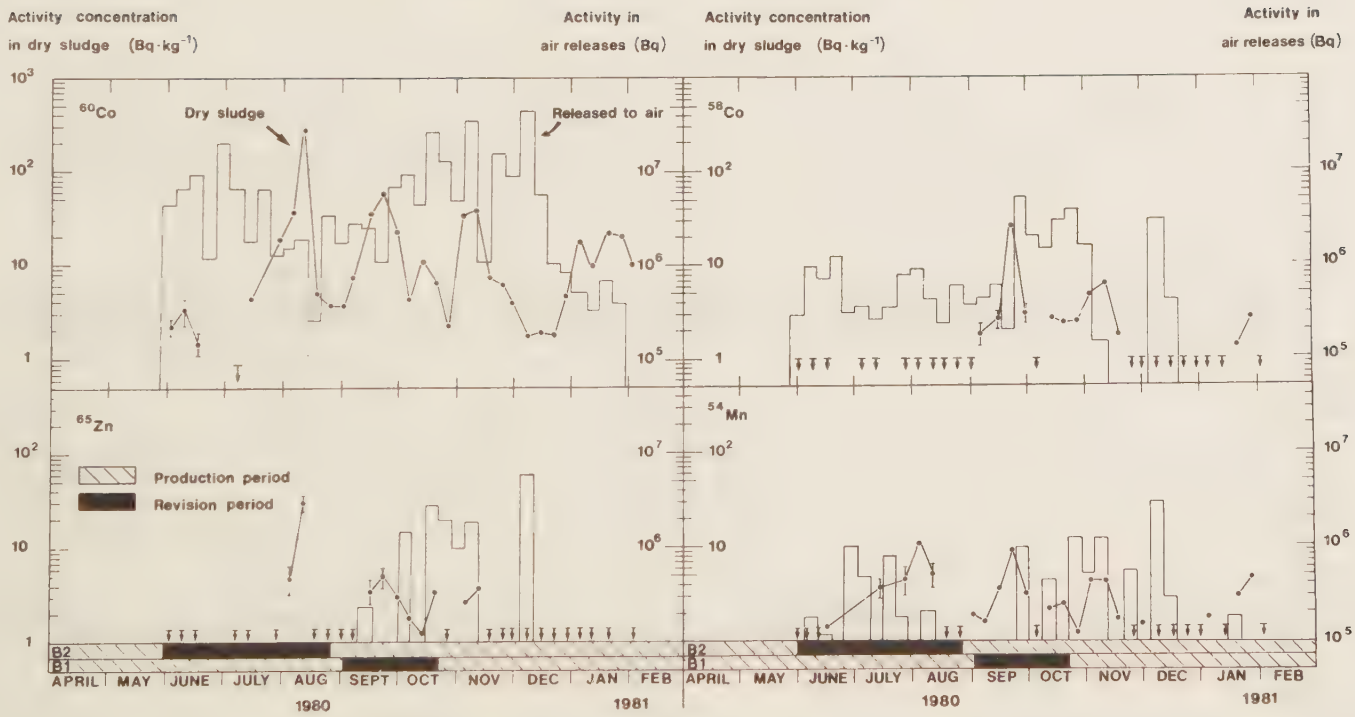

FIGURE 3 The activity concentration of ^{60}Co, ^{58}Co, ^{65}Zn and ^{54}Mn in dried samples of final sludge collected at Borgeby between April 1980 and February 1981. The reported weekly releases to the air from Barsebäck power station (5.8 km west of Borgeby) are also given. The production and revision periods for the reactors (B1 and B2) are shown in the bottom of the figure.

sequence $^{60}Co > ^{65}Zn > ^{58}Co \cong ^{54}Mn$. This squence is also valid for the activity of the reported controlled releases to the air from the power stations (OKG; Vattenfall; Sydkraft, 1980-1981). Occasionally ^{125}Sb, $^{110}Ag^{m}$, ^{57}Co and ^{51}Cr have also been detected in the sludge samples. These radionuclides are also present in the controlled releases to the air from the power stations. Detailed local wind-direction and speed data from the three investigated areas during the sampling time were not available. The lack of these data makes it hard to compare, in detail, the weekly controlled reported releases to the air from the power stations with the activity concentration found in the sludge. Figures 1, 2 and 3 show that small amounts of ^{60}Co are released to the air and that ^{60}Co is detected in all sludge samples collected at Oskarshamn, Bua, and Borgeby, with only one exception at Borgeby in the beginning of July 1980. The figures also show that high releases of ^{58}Co are followed by increases in the activity concentration in the sludge and that there is an increase in the measured activity of all four activation products in the sludge during the revision periods for the boiling water reactors O1, O2, R1, B1 and B2. Figure 2 shows that no increase in the activity concentration in the sludge could be measured during the revision of the pressurized reactor, R2, at Ringhals. A difference between the two reactor types is that the ventilation air from R2 at Ringhals is filtered before being released to the air which is not the case for the other reactors.

Table 1 shows the estimated yearly "outputs" of ^{60}Co and

TABLE 1. The reported yearly releases of ^{60}Co and ^{65}Zn to air and water from the power stations together with the estimated total activity in the final sludge which is produced during the same year.

Power station	Year	Reported yearly release (MBq)				Collecting site for sludge	Distance to the power station (km)	Estimated drained area (km^2)	Yearly "outputs" via sludge (MBq)	
		air		water						
		^{60}Co	^{65}Zn	^{60}Co	^{65}Zn				^{60}Co	^{65}Zn
Simpevarp	1978	60	+	+	+	Oskarshamn	21	2.8	6.7	2.3
Simpevarp	1979	+	+	+	+	Oskarshamn	21	2.8	6.1	2.0
Simpevarp	1980	23	14	28000	23000	Oskarshamn	21	2.8	6.5	2.2
Ringhals	1980	150	101	92000	42000	Bua	2.4	0.3	7.5	1.3
Barsebäck	1980	364	18	37000	5800	Borgeby	3-10	1.0	8.6	0.74

+ Data not available

^{65}Zn in sludge from Oskarsham, Bua and Borgeby sewage treatment plants together with the reported yearly releases from the nuclear power stations both to the air and water. The table shows that the yearly "outputs" of the radionuclides via the sludge from Oskarshamn waste water plant is about the same during 1978, 1979 and 1980 and is, from this single waste water treatment plant, as high as about 25% of the total reported releases to the air from Simpevarp. If the yearly "outputs" of ^{60}Co and ^{65}Zn in Table 1 are divided by the estimated drained areas the deposition of ^{60}Co during 1980 was 2.3, 25 and 8.6 Bqm^{-2} at Oskarshamn, Bua and Borgeby respectively and for ^{65}Zn, 0.8, 4.3 and 0.74 Bqm^{-2} respectively. It must be stressed that there is a difference in distance between the sources for the radionuclides and the various treatment plants. There is also a difference in the yearly emitted activity from the three power stations. These yearly deposition figures for the activation products may be compared with the yearly deposition of ^{7}Be, 700 Bqm^{-2}. This indicates that the deposition of the detected activation products even in the nearest surroundings of the power stations is comparatively small.

Figure 4 shows the activity ratio $^{58}Co/^{60}Co$ in sludge samples from the various treatment plants and in reported releases to the air from the power stations. For the Barsebäck area the agreement between the ratio in sludge and in reported releases to the air is rather good. This indicates that the "age" of the cobalt isotopes in the sludge samples is short compared with the physical half-life of ^{58}Co (70.8d). The limited information for the

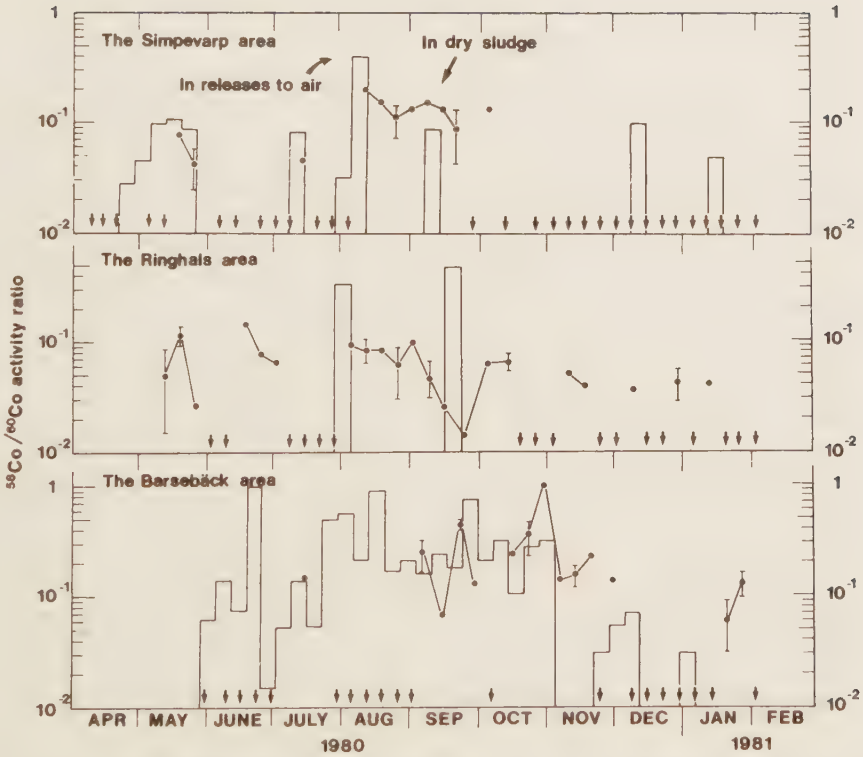

FIGURE 4 The activity ratio $^{58}Co/^{60}Co$ in reported releases to the air from the nuclear power stations and in samples of final sludge from sewage plants in their surroundings.

Simpevarp and the Ringhals areas makes such an estimation of the "age" uncertain.

The results of the analysis of "entrance sludge" are presented in Figure 5, where the activity concentration of ^{131}I, ^{75}Se, ^{60}Co and ^{7}Be and also the amount of precipitation are shown. This "entrance sludge" shows, with better time resolution than the "final sludge", the temporal variation of the activity concentration in the incoming waste water. Figure 5 shows that the activity concentration of ^{131}I and ^{75}Se are unaffected by the precipitation, which may be explained by the fact that these radionuclides are used in hospitals and enters the sewage systems via excreta from treated patients, (Mattsson et al., 1977; Erlandsson & Mattsson, 1978; Prichard et al., 1981). The activity concentration of ^{7}Be is almost doubled soon after the onset of rain. This radionuclide is continuosly produced in the atmosphere (Lal et al., 1967) and is washed out from the air by the rain. Before the start of the rain the activity concentration of ^{60}Co was below the minimum detectable limit, which was $2 Bqkg^{-1}$ (d.wt). After about 2 mm of rain it reaches a peak value of about 55 $Bqkg^{-1}$ (d.wt). and then the concentration falls off rapidly. The most likely explanation of this time variation pattern is that the ^{60}Co activity accumulates on the ground and is then washed out by the rain. This sampling has been repeated three times and has shown the same time variation pattern as shown in Figure 5.

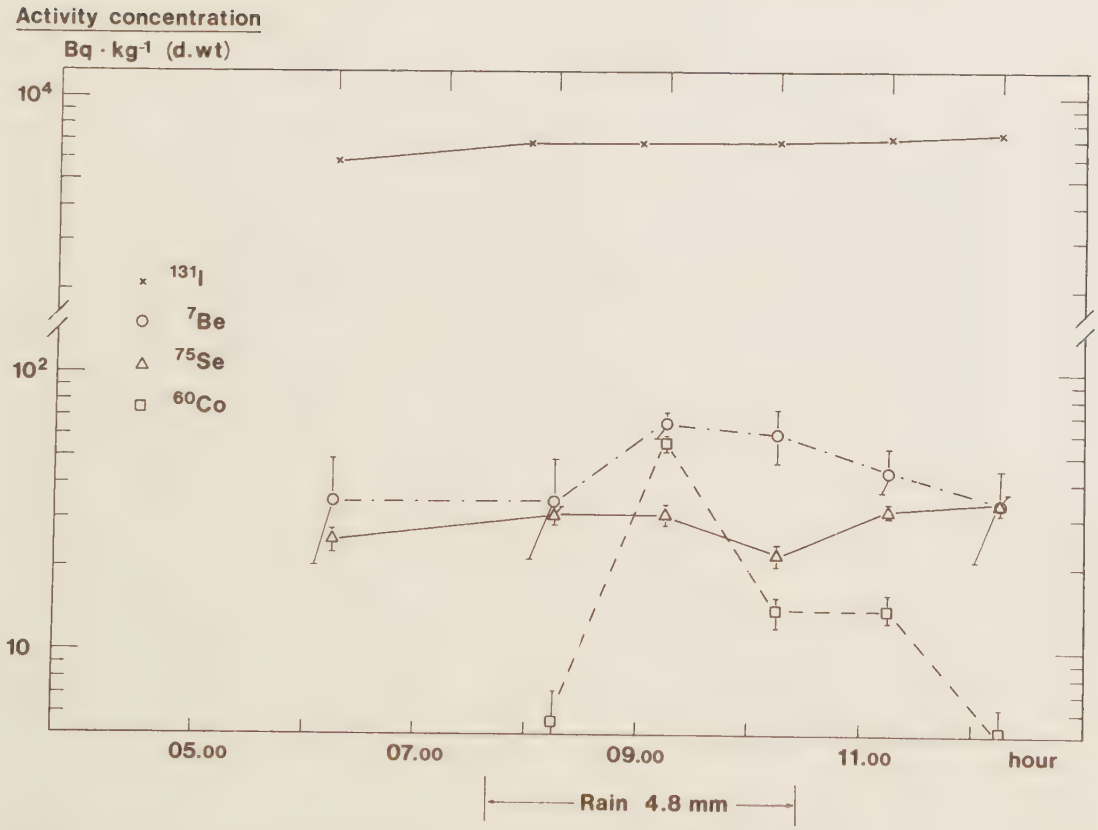

FIGURE 5 The activity concentration of ^{60}Co, ^{7}Be, ^{131}I and ^{75}Se in dried samples of entrance sludge collected at Borgeby sewage plant during a rain shower, 1980-10-02. The amount of rain is given in the bottom of the figure.

Activation products in lichen and soil

Table 2 shows the content of ^{60}Co , ^{65}Zn, and ^{7}Be per unit area in both lichen and the underlying soil, down to a depth of 5 cm, in the surroundings of the Simpevarp nuclear power station. The content of ^{60}Co is considerably higher in the underlying soil than in the lichen carpet. This is contradictory to the behaviour of most gamma-emitting fall-out products (Mattsson, 1975), which show a slow decrease, following a power function, in activity concentration by depth down through the lichen carpet and into the soil. It is therefore difficult to use lichen as a long-term indicator for ^{60}Co released from nuclear power stations. For short-term qualitative or comparative measurements it may, however, be of some value (Ingemansson et al., 1981).

Activation products in ground level air

Ground level air was sampled during five weeks in December 1980 and January 1981. The single radionuclide which normally gives the highest activity concentration in surface air filters is always ^{7}Be (up to 4 $mBqm^{-3}$). Some times other radionuclides from nuclear weapons tests also showed comparatively high activity concentration. These concentrations and also their variations with time are in good agreement with results from measurements made at Ljungbyhed, 42 km north-east of Barsebäck (ELBA, 1980-1981).

TABLE 2. The estimated area content of ^{60}Co, ^{65}Zn and ^{7}Be in three 5 m^2 samples of lichen (Cladonia alpestris) and two 0.5 m^2 samples of underlying soil down to a depth of 5 cm collected in the surroundings of Simpevarp nuclear power station.

Collecting date	Distance to the power plant (km)	Direction from the power plant	Area content Bq·m^{-2}					
			^{60}Co		^{65}Zn		^{7}Be	
			Lichen	Soil	Lichen	Soil	Lichen	Soil
79-11-17	6.2	SW	0.16		0.089		93	
80-09-22	1.1	W	0.93	2.5	0.092	<0.3	146	<16
80-10-10	3.8	SSW	0.14	2.0	0.050	<0.3	174	<16

During four of the five weeks no detectable amounts of ^{60}Co, $<0.4\ \mu Bqm^{-3}$, were found in the ground level air. During these weeks the wind was not directed from the power station to the air sampling device.During one sampling period, 81.01.27-81.02.04 the concentration of ^{60}Co in ground level air was well above the detection limit ($3.5 \pm 0.3\ \mu Bqm^{-3}$) at Borgeby. During the same period it was less than $0.3\ \mu Bqm^{-3}$ at Ljungbyhed (Arntsing, 1981). During the actual week there was a reported release from one of the reactors at Barsebäck. The mean release rate was 33 Bqs^{-1} (Sydkraft, 1981). The wind direction was also such that the plume from the chimneys of the power station was directed (within $\pm 5^{o}$) towards the sampling device for 20% of the sampling time.

An estimation of the average concentration of ^{60}Co in ground level air at different distances in the wind direction from the source of release can be made by using the well-known diffusion-equation (Slade, 1968)

$$\chi(x,y,z,h) = \frac{Q'}{2\pi\ \sigma_y\ \sigma_z\ u} \exp\{-(\frac{y^2}{2\sigma_y^2} + \frac{h^2}{2\sigma_z^2})\}$$

where:

h is the effective height of the chimney (100m)

Q' is the emmission rate from a point source (33 Bqs^{-1})

u is the average wind speed (5 ms^{-1}) for the period

(x,y,z), is the position in a Cartesian coordinate system where the x-axis is in the direction of the mean horizontal wind, the y-axis is crosswind, and the z-axis is vertical (m)

σ_y, σ_z are the standard deviations of the distribution of activity in the plume in the y- and z-directions (σ_y=340m and σ_z =100m for x=5.8km)

χ(x,y,z,h) is the time average value of the concentration (Bqm^{-3})

If no reflection from the ground is asssumed and with the parameter values above, the calculated dilution factor ($\chi(x,y,z,h)/Q'$) for Borgeby (x=5800m, y=0m, z=0m, h=100m) is $0.57.10^{-6}$ which gives an activity concentration in air at Borgeby of 3.8 μBqm^{-3} which is in good agreement with the measured value, which was 3.5 ± 0.3 μBqm^{-3}.

The activity ratios $^{60}Co/^{7}Be$ and $^{65}Zn/^{60}Co$ in different species

The activity concentration for a specific radionuclide in dry sludge, collected at different sewage plants can vary by two orders of magnitude. This difference may depend on the various distances from the source and on different proportions of rain water in the incoming water. It has been shown that these differences can to some degree be eliminated by dividing the activity concentration of the

radionuclide of interest by that of ^{7}Be (Mattsson et al., 1979; Ingemansson et al., 1981). The activity ratio $^{60}Co/^{7}Be$ is about 0.3 in sludge samples from Borgeby but in ground level air sampled at the same station the ratio is lower than 0.0025. In lichen and surface soil it is about 0.02. Possible explanations for these differences can be that ^{7}Be and ^{60}Co have different deposition mechanisms. The ^{60}Co isotope might also be more effectively washed out from the run-off areas of the sewage treatment plants and also more effectively retained in the sludge than the beryllium.

The activity ratio $^{65}Zn/^{60}Co$ is shown in Table 3. The ratio in the sludge is about the same during 1978, 1979 and 1980 at Oskarsham, near Simpevarp, and there is no significant difference in the activity ratio for sludge and lichen. The $^{65}Zn/^{60}Co$ activity ratio in the sludge and in reported releases to the air and water are of the same order of magnitude for the three investigated areas. This indicates a similar behaviour of ^{60}Co and ^{65}Zn on their way from the reactor to the sewage sludge.

SUMMARY AND CONCLUSION

Sewage sludge from waste treatment plants has been shown to be a very sensitive and convenient indicator for activation products released to the air from the three Swedish power stations at Simpevarp, Ringhals and Barsebäck. Weekly sampling of sludge during an eight month period at sewage plants in the surroundings of the power

TABLE 3. The activity ratio $^{65}Zn/^{60}Co$ in sludge collected at Oskarshamn, Bua and Borgeby, as well as in reported releases to the air and water from Simpevarp, Ringhals and Barsebäck and in lichen collected near Simpevarp.

Area	Year	Activity ratio $^{65}Zn/^{60}Co$ in reported releases		sludge	lichen
		air	water		
Simpevarp	1978	-	-	[illegible]	
Simpevarp	1979	-	-	[illegible]	[illegible]
Simpevarp	1980	0.61	[illegible]	[illegible]	[illegible] [a]
Ringhals	1980	0.67	[illegible]	[illegible]	
Barsebäck	1980	0.049	[illegible]	[illegible]	

a) Average of two samples.

stations shows that the observed radionuclides can be ordered in the following sequence after decreasing activity concentration $^{60}Co > {}^{65}Zn > {}^{58}Co \cong {}^{54}Mn$. This sequence is also valid for the reported controlled releases to the air from the power stations. There is also agreement between the time variation of the activity concentration in the sludge and in reported releases to the air from the power stations. Sludge samples collected during the first step of a sewage plant better reflect the time variation of the activity concentration in the incoming waste water than the sludge samples which are taken in the final step . Such entrance sludge samples collected before, during and after rain, show that the activity concentration of ^{75}Se and ^{131}I reaching the sewage system via patients, are unaffected by the rain, while the activity concentration of ^{60}Co rises from a non detectable level to 55 $Bqkg^{-1}$ with the first rain run-off that reaches the sewage plant and than falls off rapidly. This is strong evidence for our hypothesis that the activation products are spread via the air. Sampling of lichen and the underlying soil in the surroundings of Simpevarp nuclear power station indicates that the ^{60}Co is badly retained in the lichen carpet compared with ^{7}Be.

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THE ^{60}CO CONCENTRATION IN GROUND LEVEL AIR AT VARIOUS DISTANCES FROM A NUCLEAR POWER STATION

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ABSTRACT

Ground level air was drawn through polystyrene air filters by means of high volume air samplers. The filters were changed weekly during a five month period. The sampling devices were in operation at various sites located between 2 and 18 km from the Swedish nuclear power station at Ringhals, 52 km south of Gothenburg. Besides global radioactive fallout the filters also contained the activation products ^{60}Co, ^{65}Zn and ^{58}Co attributed to airborne releases from the power station.

For shorter distances (<6 km) the concentration of ^{60}Co in

ground level air is in quite good agreement with values predicted by a Gaussian dispersion model. For distances greater than 6 km, however, the experimental data are in better agreement with the decreasing power function, earlier suggested in connection with our studies of the variation of ^{60}Co concentration in sewage sludge, collected at various distances from a nuclear power plant.

INTRODUCTION

During the normal operation of nuclear power reactors the airborne radioactive releases through tall chimneys are dominated by inert gases, especially radioisotopes of Xe and Kr. The release of these inert gases is continuously monitored in the chimneys of the power plant. Their atmospheric dispersion has been subject to many theoretical calculations but detailed measurements in the surroundings of nuclear reactors are very few (Hsia et al., 1976). Such measurements have however been carried out around reprocessing plants (Gogolak et al., 1981; Pendergast , 1979; Telegades et al., 1978), and showed good agreement with the results of theoretical models (Slade, 1968).

Small amounts of activation products ^{60}Co, ^{65}Zn, ^{54}Mn and ^{58}Co have also been found in the terrestrial environments of some Swedish nuclear power stations (Ingemansson et al.,1981; 1982).

The purpose of this investigation was to study the

variation by distance from a nuclear power station (Ringhals, Sweden) of the activity concentration in the ground level air of different activation products, and to compare this variation with that earlier found by measurements on sludge from sewage treatment plants.

MATERIAL AND METHODS

All sampling was carried out in the surroundings of Ringhals nuclear power station, 19 km north of Varberg and 52 km south of Gothenburg on the Swedish west coast, see Figure 1. The power station has one boiling-water reactor (R1) and two pressurized water reactors (R2 and R3) with thermal effects of 2270, 2440 and 2780 MW respectively. R1 and R2 became operational in 1975 and R3 in 1980. The off-gases from the boiling water reactor (R1) are delayed about 35 minutes before they are released to the atmosphere through a 110 m high chimney together with untreated ventilation air. The total release rate is about $110 m^3.s^{-1}$. The off-gases from the pressurized reactors (R2 and R3) are normally delayed 1-45 days, depending on the drift situation. They, together with the ventilation air, are released to the atmosphere through a 45 m high chimney, after passing a HEPA (High Efficiency Particulate Air) filter. The total release rate of gases from each of these two reactors is 30-50 $m^3.s^{-1}$

Ground level air was drawn through 0.57 m x 0.57 m polystyrene air filters (Microsorban) by high volume centrifugal pumps having pumping capacities of 800 $m^3.h^{-1}$.

FIGURE 1. Map showing the locations of the Swedish nuclear power stations at Ringhals, Barsebäck and Simpevarp. The sampling sites for ground level air (+) in the surroundings of Ringhals power station are also shown together with those at Gothenburg and Ljungbyhed.

TABLE 1 Collecting sites for ground level air in the surroundings of Ringhals nuclear power station. Distances between collecting sites and power station, directions to the collecting sites relative to north from the power station and collecting dates.

Collecting site	Distance from Ringhals (km)	Direction degrees	Collecting periods 1981
Bua	1.7	160	3/8-7/9
Lahall	4.4	135	3/8-14/12
Åskloster	7.9	135	28/9-9/11
Varberg	18	155	9/11-14/12

The air velocity through the filters was 0.7 $m.s^{-1}$ and the pressure fall over the filter 6 kPa. The thickness was 1.55 mm and the surface weight of the pure filter 21.7 $mg.cm^{-2}$. The collecting efficiency of radioactive global fallout products at an intake velocity between 0.50 and 2.0 $m.s^{-1}$ has been determined to be 99.9% (Suschny, 1968). Because only two mobile air sampling devices were available, one was continuously in operation at Lahall, 4.4 km SSE of Ringhals, see Figure 1, and the other was first placed at Bua, then at Åskloster and finally at Varberg. The filters were changed weekly on the same day at the two collecting sites. Table 1 shows the distances from the power station to the sites for ground level air sampling and the directions to the collecting sites relative to the power station together with sampling dates.

The filters were pressed and packed in 180-ml plastic tubs, the density being about 0.4 $g.cm^{-3}$. The measurements were then carried out using Ge(Li)-detectors of volumes of about 100 cm^3, placed in 10 cm thick lead caves. The counting times varied between 48 and 72 hours per sample. The efficiences of the detectors were carefully determined using 180-ml plastic tubs containing sawdust, sugar or sand with densities of 0.19, 0.93 and 1.57 $g.cm^{-3}$ respectively, mixed with ^{152}Eu of known activity. The filters were normaly analysed within one week of collection. The activity concentration in the air filters has been corrected for physical decay up till the middle of the sampling period.

RESULTS AND DISCUSSION

In Figure 2 is shown a pulse height distribution from an air filter used at Lahall between 7/9-14/9, 1981. Peaks originating from man-made local, and global fallout products together with those from naturally occuring ^{40}K and ^{7}Be are marked with their respective energies. Gamma-ray peaks from the decay products of ^{238}U and ^{232}Th, which are present both in theair filters and in the detector background are marked with a plus sign (+). The activation products ^{60}Co, ^{65}Zn and ^{58}Co, attributed to the airborne releases from the power station have also been detected in the air filters. ^{60}Co is present in almost all filters from Bua, Lahall, and Åskloster, but has not been found in those from Varberg. ^{65}Zn and ^{58}Co appear occassionally in the filters. Their activity concentration is small, 5 to 10% of that of ^{60}Co. It is therefore not possible to use them continuously for quantitative studies. The contribution of ^{54}Mn from the power station to the air is negligible compared with ^{54}Mn from global fallout.

It is difficult to obtain an accurate absolute measure of the amount of air drawn through the filters during field conditions. We have therefore used the activity concentration of ^{7}Be in the air filters as a normalisation factor because its concentration in air is almost constant over an area as large as the one of interest (Wolff et al., 1979; De Geer et al., 1978). The weekly activity ratios $^{60}Co/^{7}Be$ have been calculated and are shown in

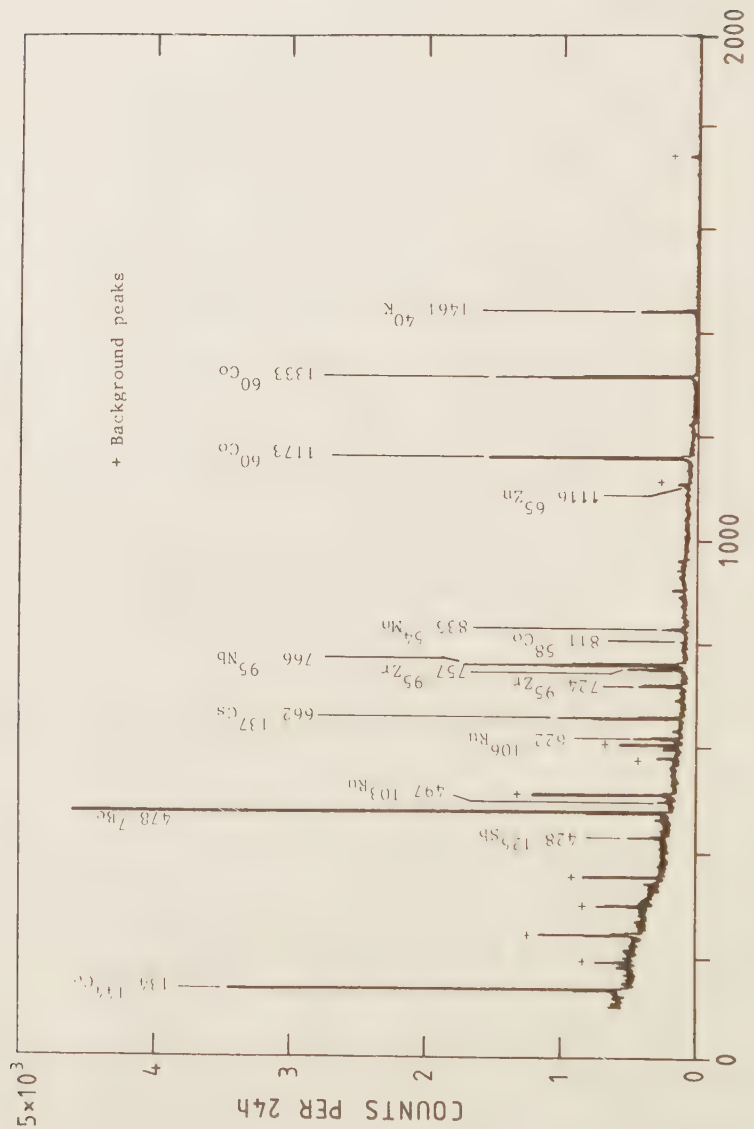

FIGURE 2. Pulse height distribution, recorded by a Ge(Li) detector, from radionuclides collected on an air filter from 7/9 to 14/9 1981, at Lahall.

TABLE 2 The activity ratio $^{60}Co/^{7}Be$ in ground level air at Bua, Lahall, Åskloster and at Varberg.

Date 1981	Bua $x10^{-4}$	Lahall $x\ 10^{-4}$	Åskloster $x10^{-4}$	Varberg $x10^{-4}$
3/8-10/8	<1.9	5.7±0.9		
10/8-17/8	5.1±0.5	19.1±1.3		
17/8-24/8	4.1±2.4	5.6±2.8		
24/8-31/8	<2.3	26±2		
31/8-7/9	4.7±0.7	21±1.3		
7/9-14/9	23±2+	330±20		
14/9-21/9		99±6		
21/9-28/9		99±6		
28/9-5/10		<0.73	<0.95	
5/10-12/10		3.6±0.5	2.2±0.7	
12/10-19/1		23±2	6.3±0.8	
19/10-26/1		<2.3		
26/10-2/11		2.8±1.4	<1.4	
2/11-9/11		4.6±1.3	<3.2	
9/11-16/11		2.7±0.8		<2.3
16/11-23/11		5.3±1		<1.8
23/11-30/11		7.3±2.7		<2.5
30/11-7/12		7.3±2.7		<2.5
7/12-14/12		3.5±0.7		<1.5

+ 7/9-9/9, device failure.

Table 2 for the different collecting sites. The variation of the ratio with time is considerable, mostly due to varying release and weather conditions. During the measurement period 3/8-14/12 1981 the activity concentration of ^{60}Co was highest at Lahall and varied between 0.3 and 100 μBqm^{-3} with a mean value of 9 μBqm^{-3}. No significant ^{60}Co activity was detected in the air filters from Varberg during the measurement period. The weekly average of the ^{60}Co activity concentration at Gothenburg and Ljungbyhed, which used the same type of sampling devices (Bernström, 1978; De Geer et al., 1978) was below the detection limit during the whole sampling period with only one exception (Ljungbyhed, 19-26/9) when it was 0.28 ± 0.06 $\mu Bq.m^{-3}$ (see Figure 1) (Arntsing, 1981).

To get a picture of the distance dependence of the activity ratios, Lahall was chosen as a normalisation point (Figure 3a). As several of these ratios only give an upper limit it is inadequate to calculate a mean value. One - sided confidence intervals (bounded from above) were therefore constructed in analogy with the sign test, having in mind a confidence level of 80-90%: because of the inequality sign for some of the individual ratios, confidence intervals so constructed will be conservative, i.e. will have confidence levels larger than those stated. With 89% confidence the ratio for Bua is <0.33, with 94% confidence the ratio for Åskloster is <0.70 and with 81% confidence the ratio for Varberg is <0.43. All these ratios are significantly less than 1. The activity ratios $X/^{7}Be$ for six other radionuclides (X:), ^{144}Ce, ^{137}Cs

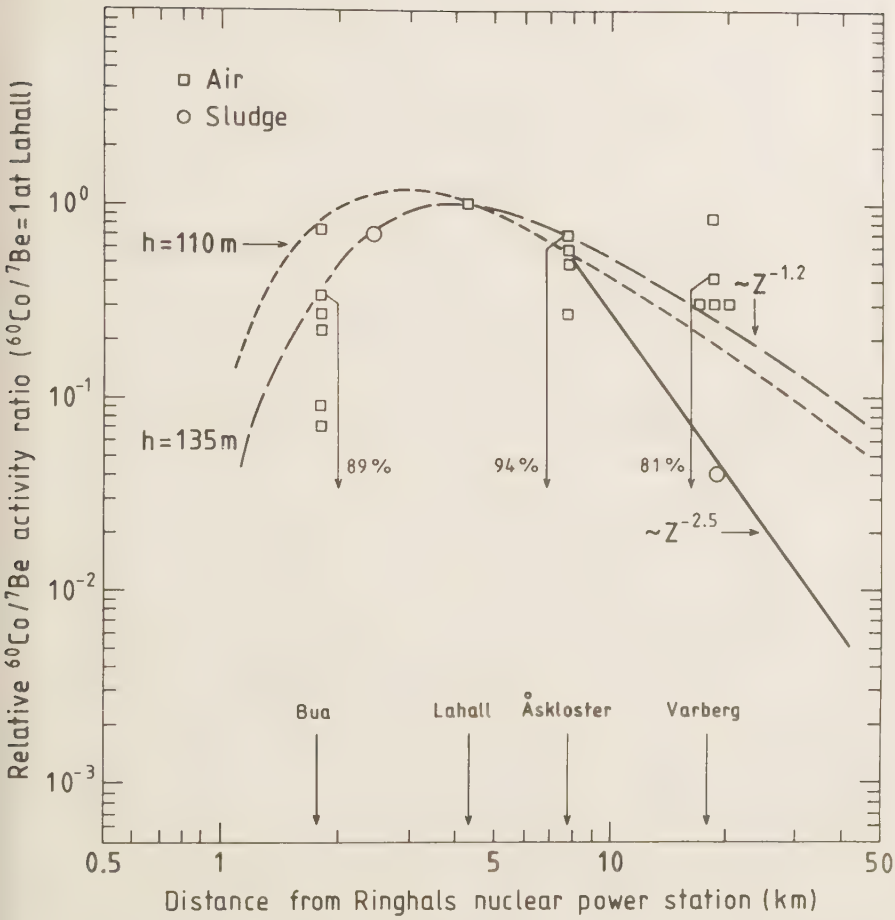

FIGURE 3a. Relative $^{60}Co/^{7}Be$ activity ratio ($^{60}Co/^{7}Be$ = 1 at Lahall) in ground level air (□) at Bua, Lahall, Åskloster and Varberg. The confidence intervals and their levels are also shown. The activity ratio $^{60}Co/^{7}Be$ in sludge (O) from Bua and Varberg are also shown together with the power function $f(z) \sim z^{-2.5}$ and a Gaussian plume model.

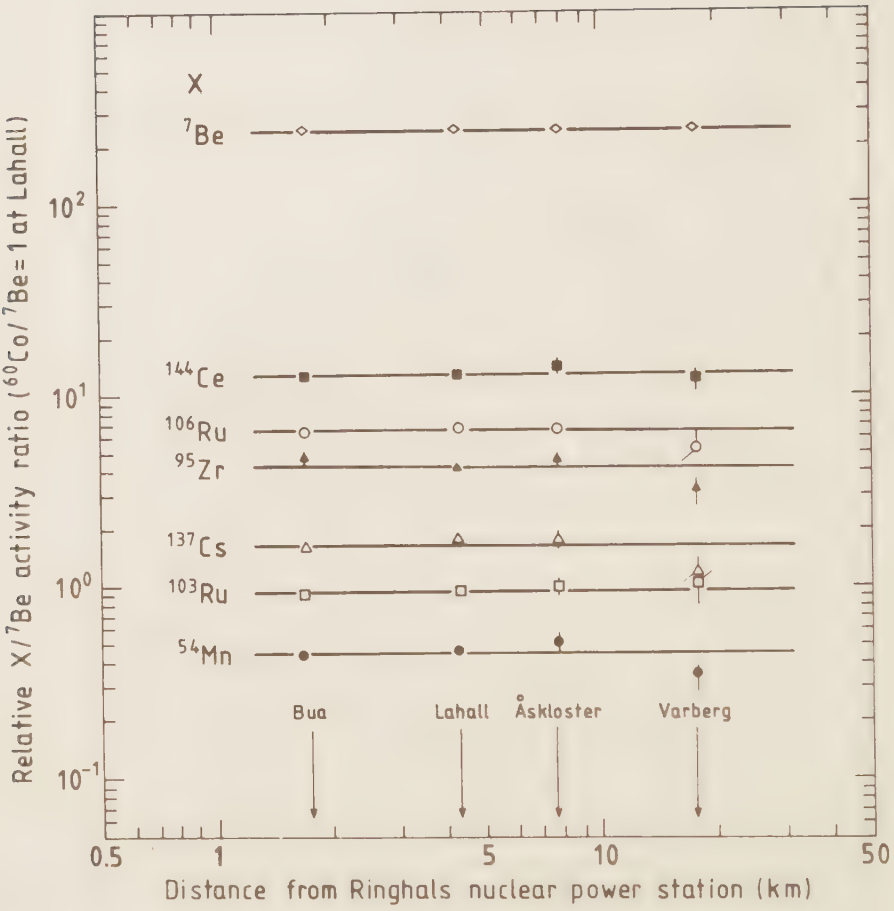

FIGURE 3b. Relative X/^{7}Be activity ratio (^{60}Co/^{7}Be = 1 at Lahall) for (X:) ^{144}Ce, ^{137}Cs, ^{106}Ru, ^{103}Ru, ^{95}Zr, ^{54}Mn and ^{7}Be in ground level air at Bua, Lahall, Askloster and Varberg.

^{106}Ru, ^{103}Ru, ^{95}Zr and ^{54}Mn were also calculated. For each sampling period the weekly ratios between the activity ratio $X/^{7}Be$ at the actual site and that at Lahall are a measure of the relative concentration of X in ground level air. These values are close to 1 for Bua, Åskloster and Varberg (Figure 3b). In order to use the same scale on the ordinate in this figure as in Figure 3a the ratios mentioned above have all been multiplied by the ratio $(X)_{mean}/(^{60}Co)_{mean}$. In this ratio $(X)_{mean}$ and $(^{60}Co)_{mean}$ denote the mean values of the activity concentration in the air filters at Lahall calculated over the whole sampling period, 3/8-14/12, for the radionuclide X and for ^{60}Co.

The pronounced distance dependence for ^{60}Co, however, indicates that the main part of this radionuclide is released from Ringhals nuclear power station. As can be seen from Figure 3b there is no significant distance dependence for ^{144}Ce, ^{137}Cs, ^{106}Ru, ^{103}Ru, ^{95}Zr and ^{54}Mn indicating that these radionuclides originate mainly from distant sources.

The experimental points obtained for the $^{60}Co/^{7}Be$ ratios in this experiment may be compared with calculations of the average long-term concentration due to releases from a point source (Slade, 1968) according to the following equation

$$\chi(x,h) = \left(\frac{2}{\pi}\right)^{\frac{1}{2}} \frac{0.01\, f\, Q\, n}{2\pi\, \sigma_z\, u\, x} \exp\{-h^2/2\sigma_z^2\} \qquad (1)$$

where $\chi(x,h)$ = concentration in ground level air ($Bq.m^{-3}$)

Q' = source release rate ($Bq.s^{-1}$)

x = distance from the emitter (m)

h = effective chimney height (m)

u = average windspeed ($m.s^{-1}$)

f = wind frequency into the sector (%)

n = numbers of sectors

$\sigma_z(x)$ = standard deviation of vertical distribution (m).

As the measured relative concentration in ground level air is a mean value over several weeks we have assumed that an "average" weather condition, Pascuill´s D-category, can be used for the calculation of σ_z.

The relation $\chi(x,h)/\chi(4500,h)$ is also presented in Figure 3a for h=110 and 135m. The releases to the air from the power station are dominated by those from the boiling water reactor (R1) (Vattenfall, 1981). The height of the chimney of this reactor is 110m. To obtain the effective release height, h, in equation (1), one dynamic and one thermal plume rise must be added to the physical height of the chimney. The total rise can be calculated to be 20-30m during a neutral weather type. This may explain the good agreement between the measured and calculated dispersion for h =135m for distances up to about 6 km from the power station.

Some measurements and calculations of the exposure rate due to inert gases from the boiling water reactor, R1, at

Ringhals have been reported (Karlberg et al., 1980). These measurements were carried out at distances between approximately 0.5 and 13 km from the power station. It was found that the calculated exposure rate was overestimated at short distances and underestimated at long distances relative to the measured values. An explanation for the discrepancy may be the use of too low a value of the effective release height, 110m, see Figure 3a.

For distances greater than 6 km from the power station the ^{60}Co concentration in the air decreases and the experimental points at Åskloster are in good agreement with theory. The experimental values at Varberg are only upper limits therefore no accurate conclusion about the agreement between theory and measurements can be drawn at this distance (18km from the power station). We have earlier found that sewage sludge is a good indicator for radionuclides in the environment (Mattsson et al., 1979, Ingemansson et al., 1981, 1982) and also that this indicator can be used to obtain a distance relation of the activity concentration of radionuclides released to the air from nuclear power stations. From measurements on sludge collected at Bua and Varberg during 1-14/8, 1981, the activity ratios $^{60}Co/^{7}Be$ were calculated to be 0.39 and 0.028 respectively, these values normalized to 0.7 at Bua are also presented in Figure 3a. The sludge value at Varberg is in good agreement with the power function $f(z) \sim z^{-2.5}$ (Ingemansson et al., 1981). This indicates that the distance relationship obtained by the Gaussian plume model tends to overestimate the activity

concentration for longer distances. There is also another fact in favour of a more rapid decrease in the activity concentration and that is that at Gothenburg, 52 km north of the power station, on no occasion during the sampling period has any measurable activity of ^{60}Co been reported (Arntsing, 1981).

SUMMARY AND CONCLUSION

The variation by distance from a nuclear power station of the ^{60}Co concentration in ground level air has been studied. The activity concentration in the air varied between 0.3 and 100 $\mu Bq.m^{-3}$ with a mean value of 9 $\mu Bq.m^{-3}$ at a collecting site 4.4 km south of the power station. At sampling sites far from the power station the ^{60}Co concentration in air did not exceed 0.3 $\mu Bq.m^{-3}$. For distances less than 6 km the measured dispersion in the air was in very good agreement with calculations using a Gaussian plume model. For distances over 6 km, however, measurements on sludge strongly indicate that the Gaussian plume model may overestimate the concentration in the air.

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COMPARATIVE STUDIES OF RADIONUCLIDES FROM GLOBAL FALLOUT AND LOCAL SOURCES IN GROUND LEVEL AIR AND SEWAGE SLUDGE

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ABSTRACT

The presence in the environment of activation products released to the air from a nuclear power station has been studied in sewage sludge and ground level air. The measured time variation of the ^{60}Co concentration in sludge and ground level air was found to be in good agreement with the reported variation in release rate to the air from the power station when the prevalent wind direction is taken into account. The ratio between the ^{7}Be normalised concentration in sludge and ground level air was studied and was found to be about 1 for radionuclides due to global fallout but less than 2.10^{-3} for ^{60}Co. Our

conclusion is that ^{60}Co spreads from a local source (a nuclear power station) and to a considerable degree is dry deposited on the ground and then washed off by rain.

INTRODUCTION

Small amounts of radionuclides are continuously released to the water (Nilsson, 1980) and the air from nuclear power reactors despite the fact that they are equipped with advanced radioactive waste handling systems. During serious accidental conditions there may be a substantial rise in the discharged activity, particularly the airborne component. When calculating the radiological impact there is a considerable need for detailed experimental knowledge of transport phenomena affecting the movement and dispersion of the released activity. In previous studies (Ingemansson, 1981 et al.; 1982; Mattsson et al., 1982) activation products released to the air from nuclear power stations were detected in samples of sewage sludge, ground level air, lichen (Cladonia alpestris) and soil. The results of these investigations showed that sewage sludge was a very sensitive and convenient indicator for radionuclides released to the air from a nuclear power station.

The purpose of the present investigation was to perform comparative studies in ground level air and sewage sludge on radionuclides from global fallout and local sources and also to study the transport of the radionuclides from the ground level air to the sludge.

MATERIAL AND METHODS

Sampling of both sludge and ground level air was carried out at Borgeby sewage treatment plant, located 5.8 km east of the Swedish nuclear power station at Barsebäck, which is situated on the west coast of southern Sweden, 18 km north of Malmö. The power station has two boiling water reactors with 580 MW electric power output each and came into operation in 1975 (B1) and 1977 (B2) respectively. Borgeby sewage plant serves mainly two communities, Löddeköpinge and Bjärred, which are situated at a distance of about 6 km in a sector 240°-300° relative the Barsebäck nuclear power station (Figure 1). The plant serves about 14,000 people, the daily incoming water volume is about 4200 m^3 and the daily output is equivalent to 1,100 kg dried sludge. The drained area, served by the sewage plant, was estimated to be 1 km^2.

The general functioning of a sewage treatment plant has been described earlier (Erlandsson et al.,1978; Ingemansson et al., 1982). Two types of sludge samples were collected, one type "entrance sludge" was collected just after the first (biological) degrading step, and the other, "final sludge" after the dehydrator. The mean residence time for the entrance sludge is about 1 hour. About 200 litres of this sludge was pumped up into a tank. The contents of the tank were carefully mixed and 2 litres of the sludge, with about 3% dry substance, was tapped from the bottom of the tank. The pump worked periodically

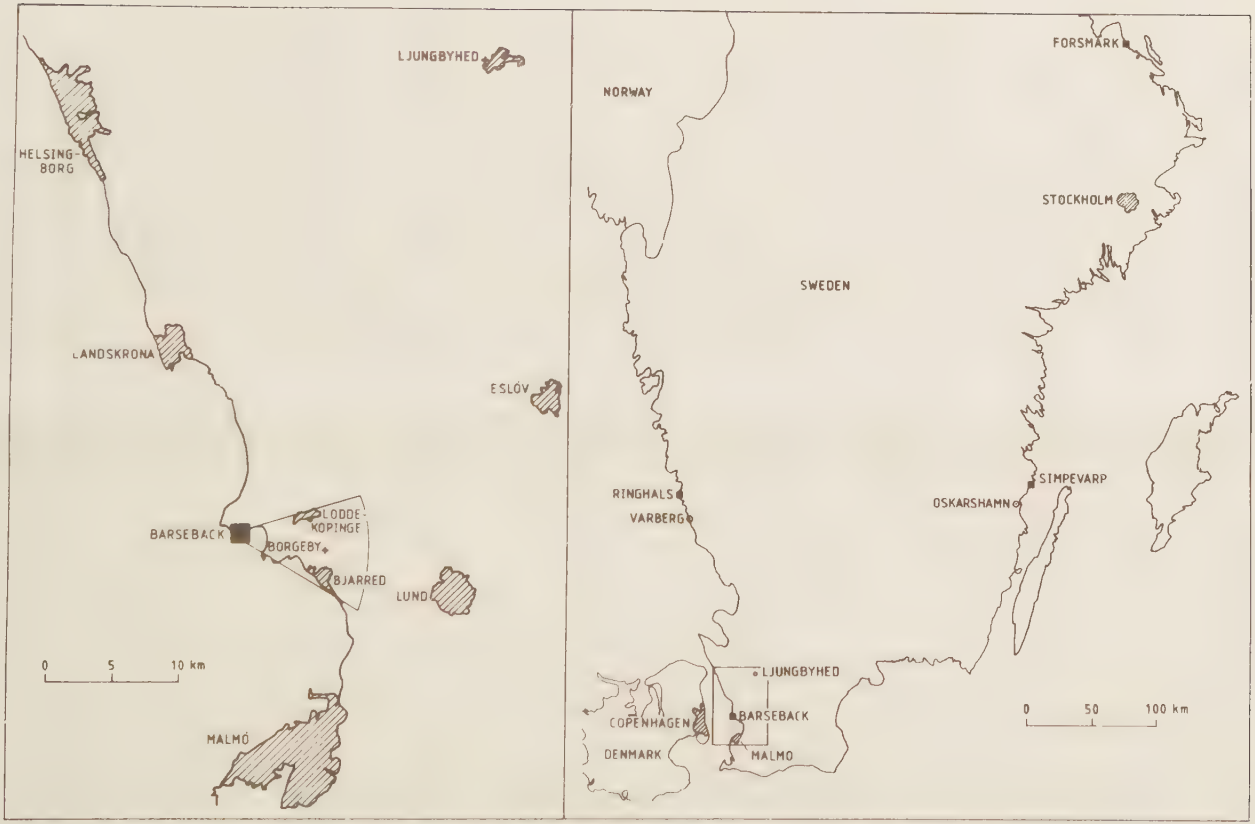

FIGURE 1. Map showing the locations of the nuclear power stations at Simpevarp, Ringhals and Barsebäck. The area served by Borgeby sewage treatment plant and the locations of the collecting sites for ground level air (+) are also shown.

(5-10 samples per hour) with an adjustable period length, whose duration was dictated by the required sampling time. The shortest sampling time was 5 minutes and the longest used in this study was 7 days. The mean residence time for "final sludge" (dry substance 14-15%) was estimated to about 5-6 days. All sludge samples were dried at 105°C for 24 hours, homogenized and packed (about 100g) in 180 ml plastic tubs. "Entrance sludge" was collected with varying sampling times 26/3-4/5, (sampling time 7 days), 14/5-18/5 (sampling time 5 minutes-4 days) and during August-September, 1981 (sampling time 5 minutes-4 days). "Final sludge" was collected about three times a week between February and September, 1981.

Atmospheric dust from ground level air was collected on 0.57 m x 0.57 m Microsorban polystyrene filters, which were placed in front of a centrifugal pump with a pumping capacity of about 800 $m^3.h^{-1}$. The filters were then packed in 180 ml plastic tubs for analysis. The air sampling was carried out weekly from February 26 to August 3, 1981.

Wind direction and speed data were obtained from the meteorological mast at the power station where measurements were made at 25 and 100 m above ground level. The precipitation data was obtained from the weather station at Lund (SMHI, 1981), located 18 km east of Barsebäck.

The gamma-ray measurements were carried out using Ge(Li)-detectors placed in lead caves with 10 cm thick walls. The

efficiencies of the detectors were determined using samples containing dried sludge or sawdust mixed with known activity of ^{152}Eu. The measured activity concentration was corrected for physical decay back to the time of collection.

RESULTS AND DISCUSSION

^{60}Co in "final sludge", ground level air and weekly reported releases to the air.

There are obviously many different mechanisms which affect the transportation of radioactive products from the source to the measuring site. In Figure 2 are shown six parameters which are of importance for the understanding of the distribution of ^{60}Co from the stacks of the nuclear power plant at Barsebäck to the air filters or the sewage treatment plant at Borgeby: a. the activity concentration of ^{60}Co in "final sludge" and b. in ground level air c. the precipitation in Lund (SMHI, 1981) d. the weekly reported releases to the air of ^{60}Co from Barsebäck nuclear power station e. "weighted" releases (weekly reported releases multiplied by the frequency of the wind direction into the 60^{o} sector in Figure 1.) and f. the revision and production periods of the two reactors. The figure shows that there is good agreement between the time variation of the "weighted" releases and that of the activity concentration of ^{60}Co in the "final sludge". During the time period June-July when the ^{60}Co concentration in the sludge is at its highest level the

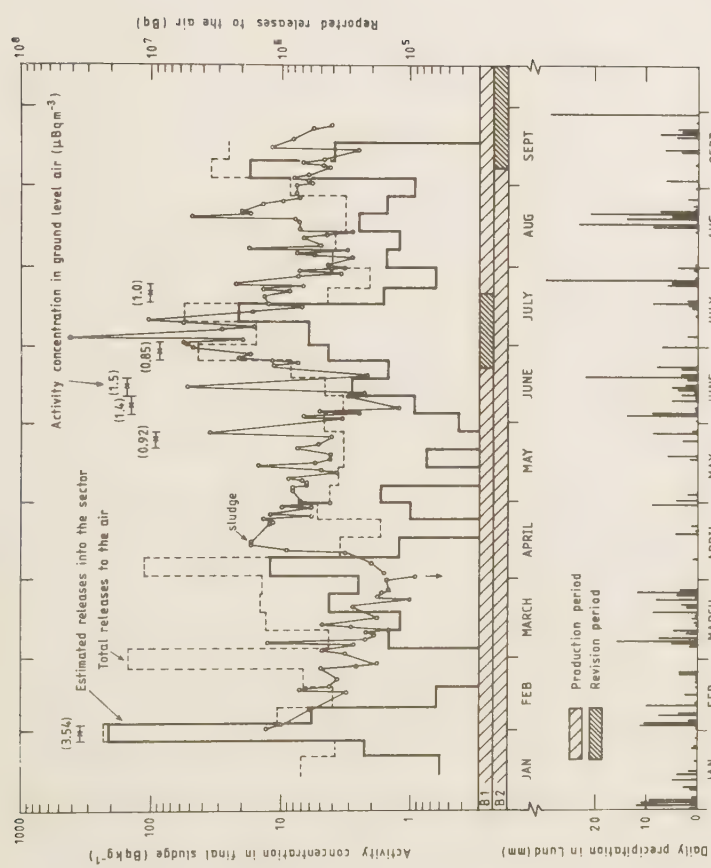

FIGURE 2. The activity concentration of ^{60}Co in dried samples of final sludge and the weekly reported releases to the air from Barsebäck nuclear power station. The measured ^{60}Co concentration in ground level air, revision and production periods for the reactors and the amount of precipitation are also shown.

radionuclide is also frequently detected in small amounts in the ground level air. During the same time period the activity concentration in the ground level air at Ljungbyhed, see Figure 1, was below 0.3 μ Bq.m^{-3} (Arntsing, 1981). This period coincided with the revision period of one of the reactors at the reactor station.

In Table 1 are shown the estimated "outputs" via the produced "final sludge" and the total and "weighted" releases of ^{60}Co, ^{65}Zn and ^{58}Co integrated over the whole sampling period, 25/1-25/9, 1981. The table shows that the relationship between the radionuclides in the "weighted" releases and in the sludge are the same. Table 1 also shows that 10% of the activity of the "weighted" releases are found in the sludge from Borgeby sewage plant, which receives rain run-off from an estimated area of about 1 km^2, situated about 5 km from the power station. Using an empirical relation (Ingemansson et al., 1981; Mattsson et al., 1982) between the distance from the power station and the ^{60}Co concentration in sludge, predicts that only 2% of the "weighted" releases should been found in sludge from Borgeby. In these calculations we have also assumed that 100% of the deposited activity is transfered from the ground to the final sludge. An assumption which is most probably an overestimation. The "weighted" releases may be subject to some uncertainties, our estimation assumes a constant release rate during the whole week. If there are considerable variations in the release rate during the week this may affect the "weighted" releases.

TABLE 1 Estimated output via sludge produced at Borgeby sewage plant together with the total reported and wind direction weighted releases to the air from Barsebäck nuclear power plant of ^{65}Zn, ^{60}Co and ^{58}Co during the period 25/1-25/9, 1981.

Radionuclide	Output via final sludge (MBq)	Activity released to the air "weighted" (MBq)	total (MBq)
^{65}Zn	0.3	3.7	6.9
^{60}Co	3.5	31.6	83.5
^{58}Co	0.3	3.6	11.0

The activity concentration in "entrance sludge".

In order to study the deposition and wash-off of various radionuclides the activity concentration in "entrance sludge" was studied during a 6 week period (Table 2 and Figure 3), during two rain showers (Figure 4) and during an 8 week period (Figure 5). Table 2 shows that the activity concentration of ^{75}Se is constant and not affected by the rain. This is to be expected beacause it is used medically and reaches the sewage system via excreta from patients. The variation patterns for ^{141}Ce, ^{103}Ru and ^{95}Zr are the same as that of ^{7}Be and Figure 3 shows that their ratios to ^{7}Be are almost constant. These ratios are about the same as those in air samples from both Borgeby and Ljungbyhed (ELBA, 1981). This indicates that these radionuclides are evenly distributed over a large area and have distant sources. Table 2 together with Figure 3 show that the variation of the activation products ^{60}Co, ^{54}Mn, ^{65}Zn and ^{58}Co differ significantly from the variation of the globally distributed radionuclides. This indicates that they may have another and perhaps more locally situated source. This sampling period was preceded by intense rain which continued into the first days of the period (Figure 2). Table 2 shows, however, that this did not affect the activity concentration of ^{60}Co and ^{54}Mn. During the second week (2-9/4) there was a substantial rise in the "input" of the activation products which was most probably caused by the slight rainfall that ended a 14 day drought period. An interesting point is that from the 28/3 up to the

TABLE 2 The activity concentration of ^{141}Ce, ^{103}Ru, ^{95}Zr, ^{75}Ce, ^{65}Zn, ^{60}Co, ^{58}Co, ^{54}Mn and ^{7}Be in dried samples of entrance sludge collected during March, April and May 1981 at Borgby sewage treatment plant. The precipitation is also given.

Date (1981)	Activity concentration in dry sludge ($Bq.kg^{-1}$)									Precipitation (mm)
	^{141}Ce	^{103}Ru	^{95}Zr	^{7}Be	^{75}Se	^{65}Zn	^{60}Co	^{58}Co	^{54}Mn	
26/3-2/4	9.9±0.4	18.1±0.6	53±1	61±4	4.0±0.3	<1	0.6±0.2	<0.6	0.9±0.2	15.5 (26-27/3)
2/4-9/4	6.5±0.4	12.4±0.6	41±1	50±4	4.2±0.3	2.4±0.7	56±1	2.0±0.4	5.2±0.4	0.9 (8-9/4)
9/4-16/4	4.7±0.9	8.0±0.8	29±2	35±6	5.6±0.6	<1	21±1	<0.6	3.5±0.6	0.0
16/4-23/4	5.8±0.6	8.6±0.7	31±2	36±4	4.3±0.4	<1	18±1	<0.6	2.9±0.4	5.9
23/4-29/4+	-	-	-	-	-	-	-	-	-	9.0 (29/4)
29/4-4/5	5.3±0.8	5.4±0.8	25±2	30±5	4.6±0.5	<1	12±1	<0.6	1.6±0.4	6.1

+ No measurements made

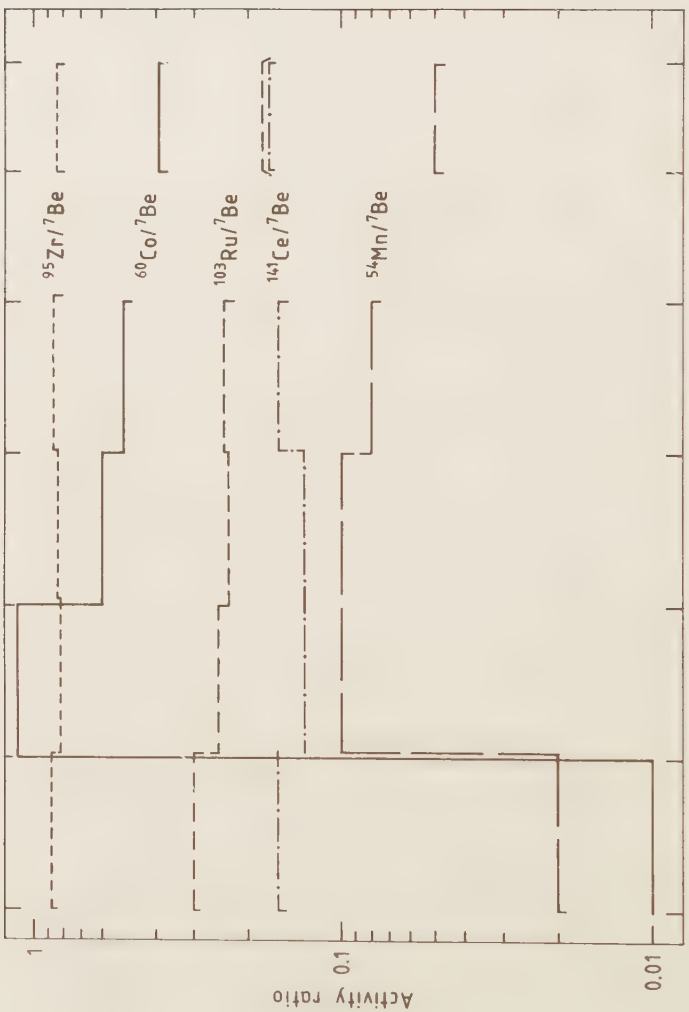

FIGURE 3. The activity ratios of ^{141}Ce, ^{103}Ru, ^{95}Zr, ^{60}Co and ^{54}Mn to ^{7}Be in weekly entrance sludge samples during March, April and May, 1981.

afternoon of the 5/4 the wind was blowing in the opposite direction to the wash-off area thus making transport of activation products to the sewage treatment plant impossible. During this week (2-9/4), the total activity in "entrance sludge" was 0.43, 0.018, 0.015 and 0.040 MBq for ^{60}Co, ^{65}Zn, ^{58}Co and ^{54}Mn respectively.

Figure 4 shows that the activity concentration of ^{60}Co in the "entrance sludge" increased from 5 to about 50 $Bq.kg^{-1}$ with the first rain run-off (only 0.9 mm) that reached the sewage plant and then fell off rapidly. The second rain shower, although as much as 3.3 mm, only accounted for a small rise from 5 to about 15 $Bq.kg^{-1}$. (The rainfall was measured at Borgeby.) The activity concentration of ^{95}Zr is almost constant in contrast to that of ^{60}Co during the sampling period.

Figure 5 shows that increased "inputs" of ^{60}Co are always correlated to rain and especially those which follow a drought period. On August 26 the activity concentration of ^{60}Co, ^{95}Zr and ^{7}Be increased and then fell off rapidly, the same variation pattern as obtained earlier (Figure 4, Ingemansson et al., 1982). Figure 5 also shows that during the whole period the activity concentration of ^{60}Co varied between about 2 and 300 $Bq.kg^{-1}$ that of ^{95}Zr between 1 and 50 $Bq.kg^{-1}$ and that of ^{7}Be between 25 and 100 $Bq.kg^{-1}$. The ^{7}Be concentration increases with increasing amount of rain.

On August 13 the "input" of ^{60}Co activity due to only 0.9

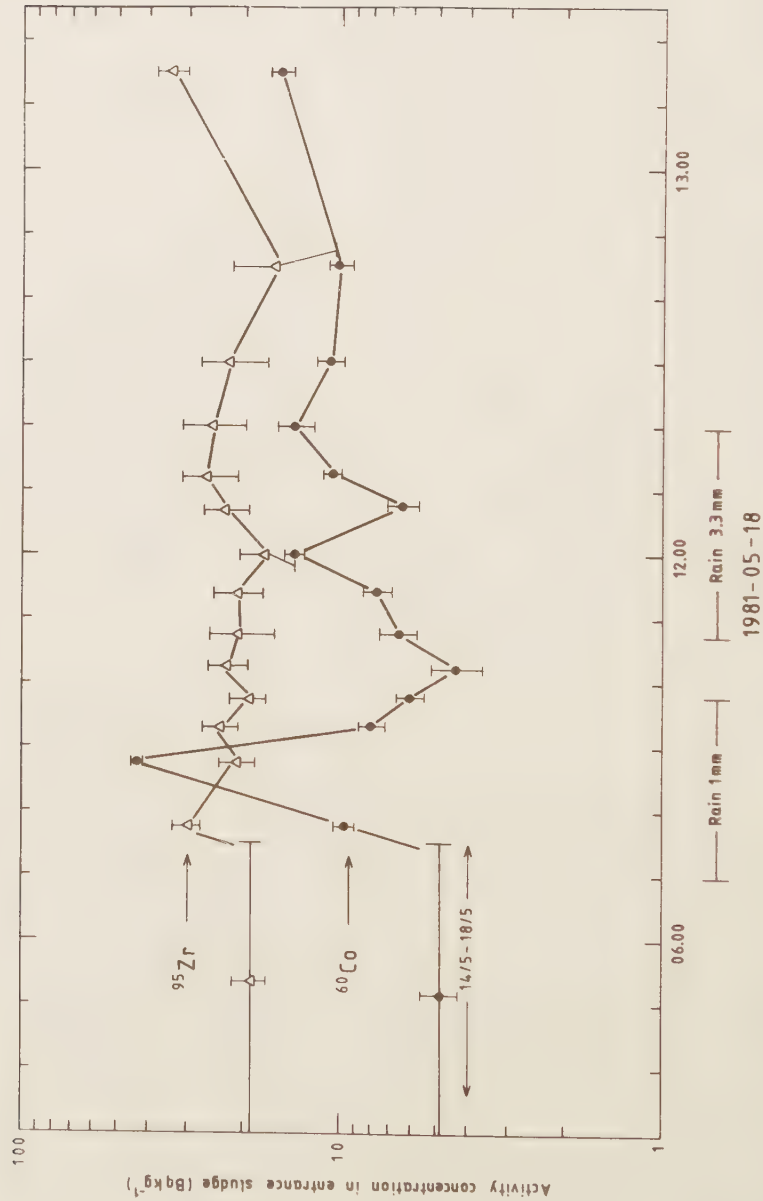

FIGURE 4. The activity concentration of ^{60}Co and ^{95}Zr in dried samples of entrance sludge collected at Borgeby sewage plant during two rainshowers 1981-05-18. The amount of rain is also given in the figure.

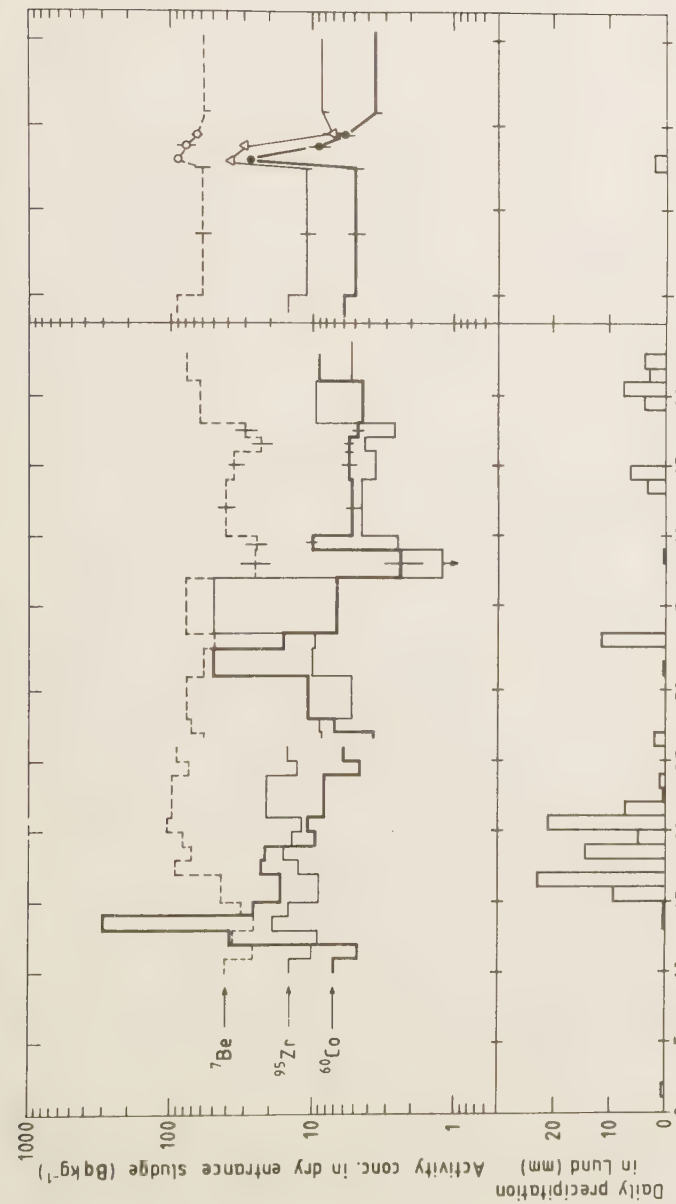

FIGURE 5. The activity concentration of ^{60}Co, ^{95}Zr and ^{7}Be in dried samples of entrance sludge collected at Borgeby sewage plant during August and September, 1981. The amount of rain is also given in the figure.

mm of rain is as high as 0.33 MBq and the total reported release of ^{60}Co to the air from the power station during 30/7-13/8 was 0.78 MBq (Sydkraft, 1981). This means that as much as 50% of the released ^{60}Co activity has been transfered to the entrance sludge. The accumulated "input" of ^{60}Co to the sewage plant during the 45 days sampling (Figure 5) can be estimated to be 0.86 MBq. During eight of these days, in connection with rain, the "input" was 0.56 MBq, which is about 65% of the total. The "input" on the 20 days with rain can be estimated to be more than 85% of the total input.

There are indications in the examples given above that even small amounts of rain have a considerable wash-off effect on ^{60}Co and especially so when the rain follows a drought period. The activity concentration in sludge is however not correlated to the amount of rain. Another explanation of this phenomena could be that the radionuclide is stored up in the sewage system during drought periods and then washed-off when rain starts to fall. Such a great effect by a small amount of rain is however not seen for globally distributed radionuclides, and studies of transport processes in the sewage system of Malmö using ^{131}I have shown that material is transported directly to the sewage treatment plant without any significant sedimentation (Erlandsson et al., 1979).

Comparison between the activity concentration measured in "entrance sludge" and "final sludge".

Table 3 shows a comparison between the activity concentration in dried samples of entrance sludge and final sludge. The table shows that the activity concentration in the entrance sludge of the global fallout radionuclides ^{144}Ce, ^{141}Ce, ^{106}Ru, ^{103}Ru, ^{95}Zr and ^{7}Be is about the same as in the final sludge. This indicates that the radionuclides are most probably attached to sedimentable material already at the beginning of the treatment process of the waste water. For ^{60}Co, however, the activity concentration is about 2.3 times higher in the entrance sludge than in the final sludge showing that entrance sludge may be a better indicator for locally released radionuclides than final sludge. One explanation for this discrepancy may be that the substantial peaks of activity with short duration in the entrance sludge (5-10 samples per hour) are almost unaffected during the treatment process and that the final sludge, sampled about three times a week, does not in a proper way reflect these peaks. This would also explain the large fluctuations in the activity concentration between consequtive samples of final sludge (Figure 2).

Relation between the activity concentration of various radionuclides in ground level air and "final sludge".

We have shown (Mattsson et al., 1979) that the activity ratio $X/^{7}Be$, where X denotes a short lived radionuclide in

TABLE 3 The activity concentration in dried samples of entrance and final sludge of ^{144}Ce, ^{141}Ce, ^{106}Ru, ^{103}Ru, ^{95}Zr, ^{60}Co, ^{54}Mn, ^{7}Be, ^{131}I and ^{75}Se and the mean ratio of the activity concentration in entrance to that in final sludge.

Nuclide	Activity concentration in dry sludge (Bq.kg^{-1})				Mean ratio entrance/final
	entrance 26/3-4/5	final^{+} 1/4-9/5	entrance 10/8-24/9	final^{+} 15/8-25/9	
^{144}Ce	15	10	-	-	1.5
^{141}Ce	6.5	4.3	-	-	1.5
^{106}Ru	7.7	6.8	-	-	1.1
^{103}Ru	11	10	-	-	1.1
^{95}Zr	36	28	13	9.6	1.3
^{60}Co	22	8.5	18	9.1	2.3
^{54}Mn	2.8	1.5	1.4	1.4	1.4
^{7}Be	42	37	60	55	1.1
$^{131}I^{++}$	183	91	1750	1350	1.7
$^{75}Se^{++}$	4.6	4.0	-	-	1.2

+ Corrected for physical decay 5 days.

++ Used in medical tests.

the global fallout, is approximately the same in sewage sludge from different places in our sampling network. The mechanisms for the transfer of a number of elements from air to rain (wet deposition) has also been shown to be similar to that of ^{7}Be (Crecelius, 1981). When this ^{7}Be normalisation is applied to sewage sludge it takes into consideration differences in the amount of precipitation and sizes of collecting areas, and when it is applied to air samples it takes into consideration differences in filter efficiency and pumping capacities.

In Figures 6-11 are shown the activity ratio $X/^{7}Be$ both in individual samples of final sludge and air filters for (X:) ^{103}Ru, ^{95}Zr, ^{65}Zn, ^{60}Co, ^{58}Co and ^{54}Mn. The differences in the $X/^{7}Be$ ratio between sludge and air is comparatively small for ^{103}Ru and ^{95}Zr and ^{54}Mn. The $X/^{7}Be$ ratio for ^{65}Zn, ^{60}Co and ^{58}Co in sludge is , however, much greater than the corresponding ratio in air. For radionuclides in the global fallout (^{144}Ce, ^{141}Ce, ^{137}Cs, ^{125}Sb, ^{106}Ru, ^{103}Ru, ^{95}Zr, ^{54}Mn as well as ^{58}Co and ^{65}Zn there is very good agreement between the $X/^{7}Be$ ratio in ground level air at Borgeby and that measured by the ELBA-group at Ljungbyhed (ELBA, 1981; Arntsing, 1981). Air and sludge ratios from Borgeby averaged over a two week period for some of these radionuclides are shown in Table 4. In this table the factor $(X/^{7}Be)_{air}/(X/^{7}Be)_{sludge}$ is also shown. This air/sludge relation has also been calculated for ^{144}Ce, ^{137}Cs, ^{125}Sb, ^{106}Ru and ^{54}Mn and is between 0.1 and 1 for most of the globally distributed radionuclides (Figure 12). Although there are few weeks with measurable

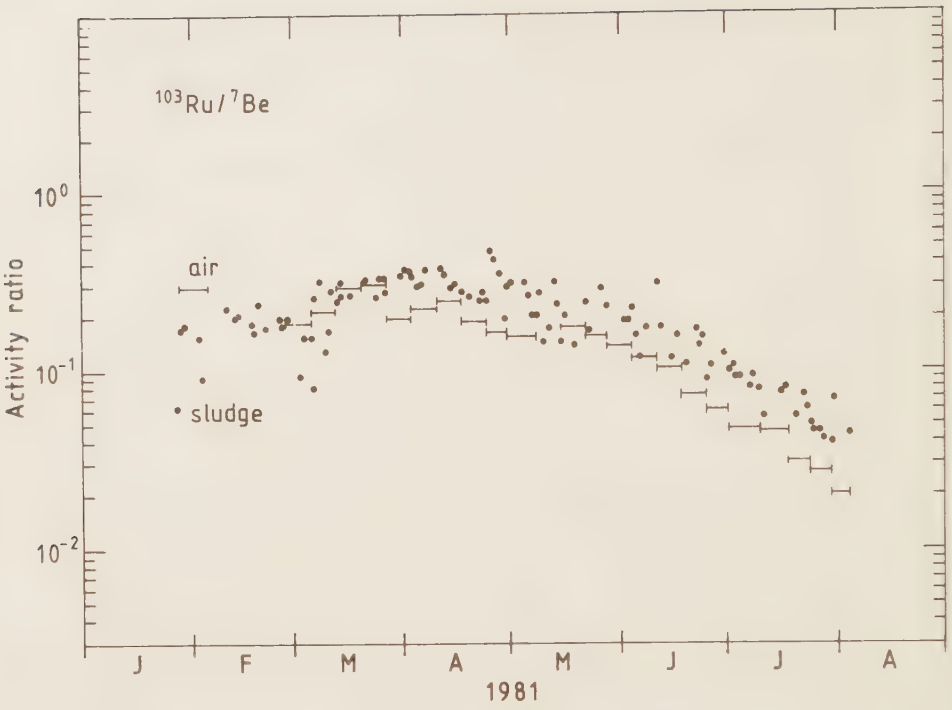

FIGURE 6. The activity ratio $^{103}Ru/^{7}Be$ in ground level air and dry final sludge.

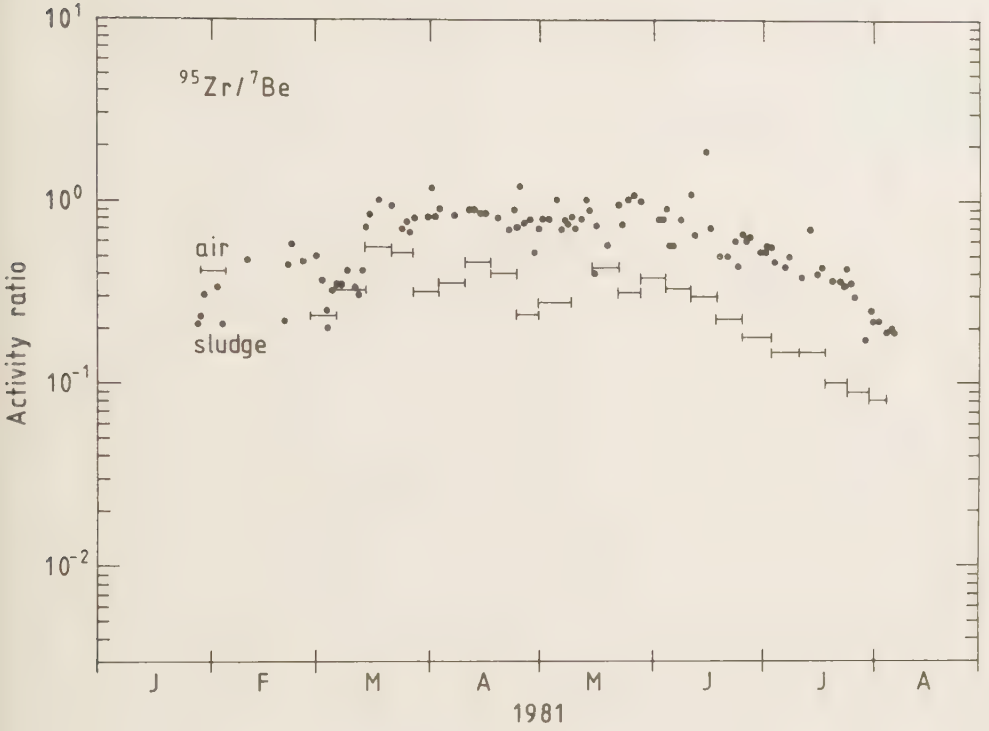

FIGURE 7. The activity ratio $^{95}Zr/^{7}Be$ in ground level air and dry final sludge.

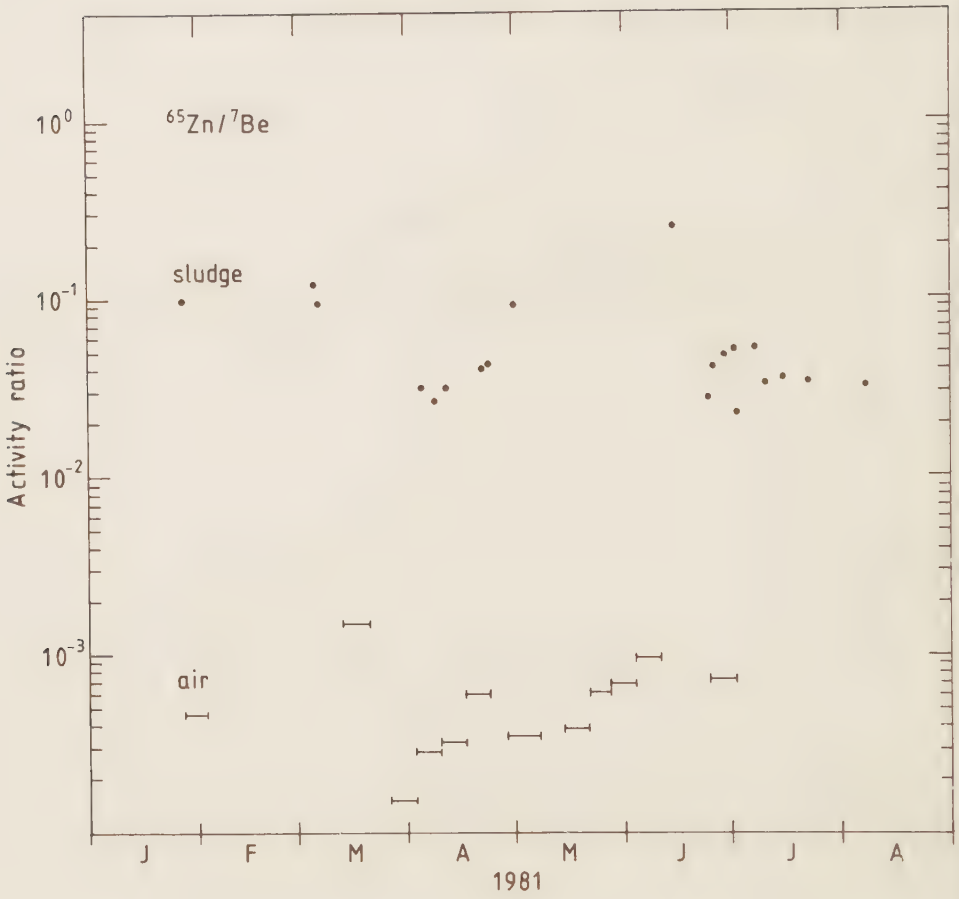

FIGURE 8. The activity ratio $^{65}Zn/^{7}Be$ in ground level air and dry final sludge.

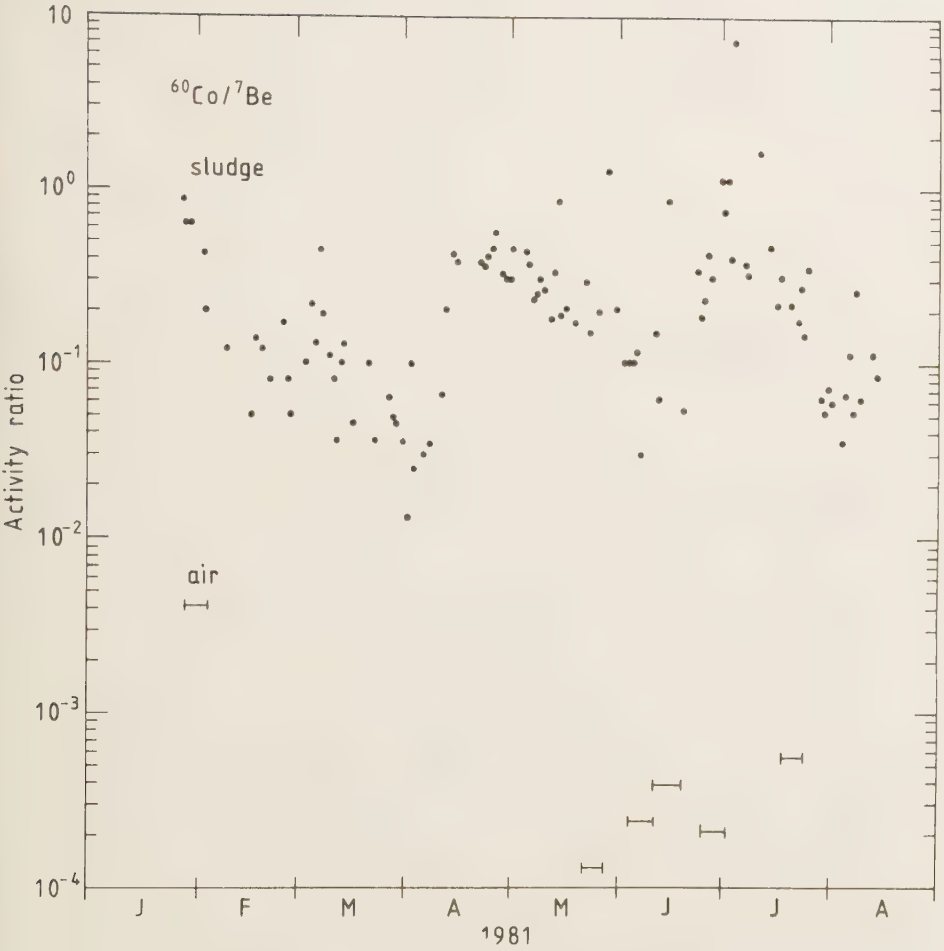

FIGURE 9. The activity ratio $^{60}Co/^{7}Be$ in ground level air and dry final sludge.

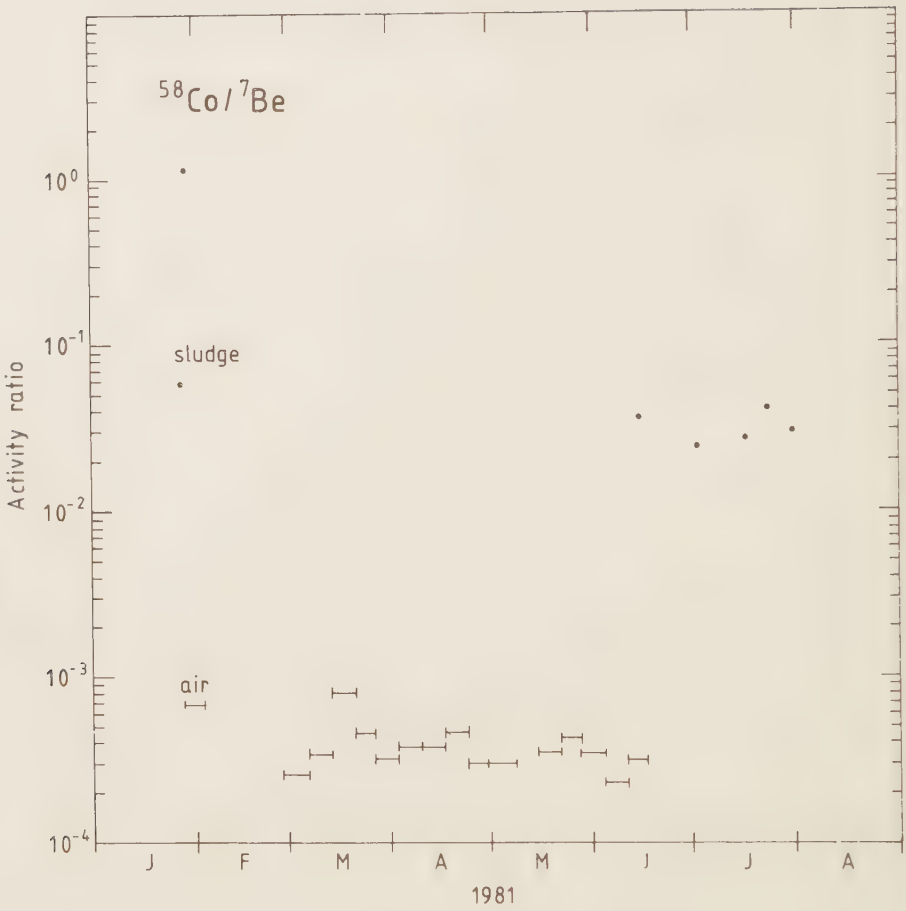

FIGURE 10. The activity ratio $^{58}Co/^{7}Be$ in ground level air and dry final sludge.

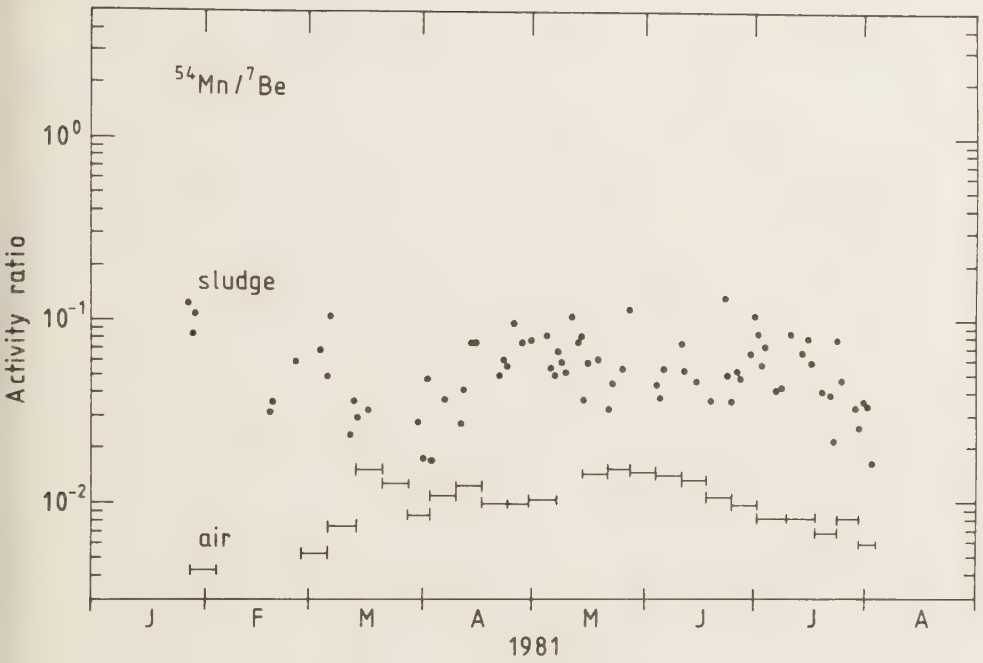

FIGURE 11. The activity ratio $^{54}Mn/^{7}Be$ in ground level air and dry final sludge.

values of ^{58}Co or ^{60}Co in both air and sludge samples (Figures 9 and 10) we have put some upper or lower limits for the air/sludge relation for these radionuclides. By using the detection limit of ^{60}Co in air we have obtained a value of the air/sludge relation which is less than 0.0017 with 97% confidence. By using the detection limit of ^{58}Co in sludge we have obtained a value of the air/sludge relation which is greater than 0.018 with 94% confidence for the period 26 February to 10 June 1981. These values were obtained by constructing one-sided confidence intervals in analogy with the sign test. For globally distributed radionuclides this relation is approximately the same at two other collecting sites (Bua and Varberg 2.4 km and 19 km south of Ringhals nuclear power station). The activity ratio of $^{60}Co/^{7}Be$ in air at Lahall and in sludge from Bua, averaged over two week periods, is shown in Table 5. The ^{60}Co activity concentration in air at Lahall is about 3 times higher than that at Bua (Mattsson et al., 1982). Therefore the $^{60}Co/^{7}Be$ value in air at Lahall has been divided by a factor 3 to get the $^{60}/^{7}Be$ value in ground level air at Bua. From the observed values a two-sided confidence interval has been constructed in analogy with the sign test. With 98% confidence the air/sludge relation in Bua lies in the interval 0.00035-0.0012. This is in good agreement with the value (<0.0017) obtained at Borgeby.

The air/sludge ratios for various radionuclides at Borgeby (Figure 12 and Table 4) show great variations, a factor of about 1000. Radionuclides which have exclusively a global

TABLE 4. The activity ratio $X/^{7}Be$ for X ^{141}Ce, ^{103}Ru, ^{95}Zr and ^{60}Co in ground level air and final sludge (Borgeby) averaged over two week periods together with the air/sludge ratios.

Date (1981)	The activity ratio $X/^{7}Be$ in								$(X/^{7}Be)air/(X/^{7}Be)$ sludge			
	ground level air				sludge+							
	^{141}Ce	^{103}Ru	^{95}Zr	^{60}Co	^{141}Ce	^{103}Ru	^{95}Zr	^{60}Co	^{141}Ce	^{103}Ru	^{95}Zr	^{60}Co
				$x10^{-4}$								$x10^{-3}$
26/2-13/3	0.21	0.20	0.29	<1.2	0.13	0.21	0.51	0.16	1.6	0.96	0.56	<0.76
13/3-26/3	0.32	0.31	0.55	<2.6	0.15	0.33	0.85	0.049	2.1	0.93	0.65	<5.3
26/3-9/4	0.18	0.22	0.68	<0.55	0.11	0.35	0.88	0.19	1.7	0.63	0.77	<0.29
9/4-23/4	0.17	0.23	0.44	<0.96	0.13	0.32	0.82	0.40	1.3	0.71	0.54	<0.24
23/4-7/5	0.12	0.17	0.26	<0.94	0.15	0.26	0.83	0.32	0.84	0.65	0.32	<0.29
14/5-27/5	0.12	0.17	0.38	<0.86	0.12	0.21	0.81	0.37	0.98	0.81	0.47	<0.23
27/5-10/6	0.082	0.13	0.36	<2.0	0.090	0.19	1.0	0.20	0.91	0.68	0.36	<1.0
10/6-24/6	0.055	0.092	0.26	<2.5	0.064	0.13	0.53	0.38	0.86	0.70	0.50	<0.66
24/6-9/7	0.031	0.055	0.17	<1.6	0.038	0. 99	0.51	1.5	0.82	0.56	0.33	<0.11
9/7-23/7	0.020	0.041	0.13	<3.5	0.037	0.060	0.37	0.21	0.53	0.68	0.35	<1.7
23/7-3/8	0.015	0.025	0.086	<2.4	0.027	0.018	0.26	0.028	0.56	1.37	0.33	<8.3
								$\bar{X}$ =	1.1	0.8	0.5	<0.0017
								1 s.d.	±0.5	±0.2	±0.2	(97%)

+Corrected for physical decay 5 days.

TABLE 5 The activity ratio $^{60}Co/^{7}Be$ in ground level air at Lahall and in final sludge from Bua, averaged over two week periods, together with the ratios divided by a factor 3. (See text.)

Date (1981)	The activity ratio $^{60}Co/^{7}Be$ air Lahall(A)	sludge Bua+(B)	1/3 A/B
3/8-17/8	12.4×10^{-4}	0.378×10^{-4}	1.1×10^{-3}
17/8-31/8	15.8 "	0.429 "	1.2 "
31/8-14/9	176 "	0.697 "	8.4 "
14/9-28/9	99 "	3.14 "	1.1 "
28/9-12/10	2.2 "	0.543 "	0.14 "
12/10-26/10	13 "	0.667 "	0.65 "
26/10-9/11	3.7 "	0.276 "	0.45 "
9/11-23/11	4.0 "	0.377 "	0.35 "
23/11-7/12	7.3 "	0.354 "	0.69 "
7/12-14/12++	3.5 "	0.325 "	0.36 "

+ Corrected for physical decay 5 days.

++ Sampling interrupted.

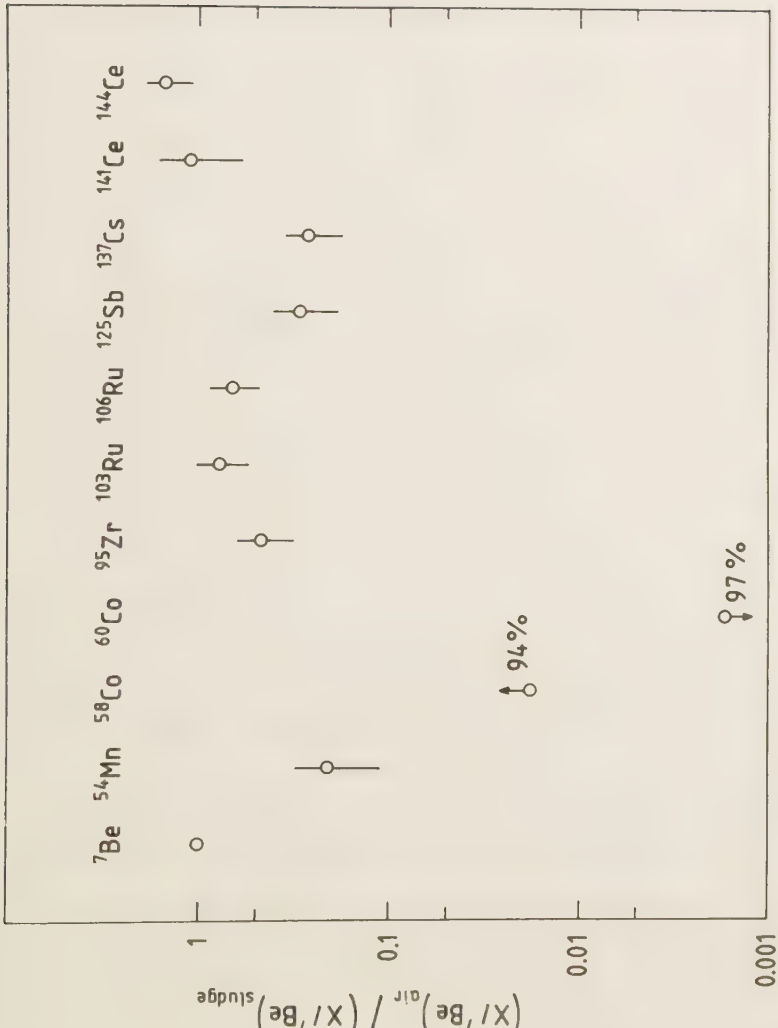

FIGURE 12. The double ratio $(X/^{7}Be)_{air}/(X/^{7}Be)_{sludge}$ for some radionuclides (X).

origin (^{144}Ce, ^{141}Ce e.t.c.) behaves like ^{7}Be. ^{60}Co differs strongly from this behaviour. One explanation of this discrepancy may be that the ^{60}Co activity comes from a local source and is therefore distributed and deposited in quite a different way than the ^{7}Be-like radio-nuclides. The difference between ^{58}Co and the ^{7}Be- like radionuclides is not so great as for ^{60}Co which may indicate that ^{58}Co is to a greater extent more globally distributed than ^{60}Co. If one assumes that there is similar behaviour for all elements in the sewage sludge process the discrepancies may be attributed to the fact that most of the ^{60}Co is released to the air from the nuclear power station, carried with the wind to the collecting area, is dry deposited and then washed off by following rain. The idea of dry deposition is supported by the wash-off pattern for ^{60}Co by rain after a drought period. Dry deposition ought to be more effective in transporting the activation products from the air to the sewage sludge than wet deposition because it is a process working all the time and not only during precipitation (it is raining 5-10% of the total time).An investigation has shown that the wind has been isotropically distributed over the sector when it has been blowing from Barsebäck into the sector. The discussion for ^{60}Co may also be valid for small fractions of the ^{54}Mn and ^{58}Co activity. The main part of these radionuclides at Borgeby, however, seems to have a global origin.

SUMMARY AND CONLUSION

Our comparative measurements of the concentration of different radionuclides in ground level air and sewage sludge have shown that the activity ratio $X/^{7}Be$ in air to that in sludge is about 1 for globally distributed radionuclides but less than 2.10^{-3} for ^{60}Co. The low value for ^{60}Co may be explained by assuming that this radionuclide is dry deposited on the ground and then washed off by rain. The idea of dry deposition is supported by the wash-off pattern for ^{60}Co by rain after a drought period. The results of our measurements strongly support the hypothesis that ^{60}Co and also other activation products are released to the air from nuclear power stations.

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